RELIABLE MOISTURE DETERMINATION IN POWER TRANSFORMERS

Von der Fakultät Informatik, Elektrotechnik und Informationstechnik der Universität Stuttgart zur Erlangung der Würde eines Doktor-Ingenieurs (Dr.-Ing.) genehmigte Abhandlung

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Jauchzet, frohlocket!
Auf preiset die Tage!
Rühmet, was heute der Höchste getan!

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Soli Deo Gloria

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Abstract

This thesis aims to provide reliable methods of assessing moisture in oil-paperinsulated power transformers. Water in power transformers causes three damaging effects: it decreases the dielectric withstand strength, accelerates cellulose aging and causes the emission of gaseous bubbles at high temperatures. Therefore knowledge about the moisture concentration in a transformer is of great importance for safe operation and for further maintenance actions.

The traditional method of moisture evaluation, oil sampling with subsequent application of an equilibrium diagram, suffers from severe errors resulting into a very poor accuracy. Similarly, the recently introduced dielectric diagnostic methods under some conditions show contradictory results. This thesis discusses and improves both moisture measurement approaches.

At first, *direct* moisture measurement techniques as Karl Fischer titration and capacitive probes were examined in order to eliminate interferences and provide a reliable basis for measurements by novel approaches.

Then the *indirect* measurement procedure using moisture equilibrium was investigated resulting in the recommended moisture assessment method for online application. Therefore moisture adsorption in cellulose (paper, pressboard) and solubility in insulation oils were theoretically described and represented as sorption isotherms and equilibrium diagrams under the special consideration of aging. Several problems led to the key recommendation of this thesis which is to use water saturation in oil *and in paper* to describe water in power transformers. All the effects of water correlate better with water saturation than with the traditionally used moisture relative to weight and it is additionally more precise measurable.

As a second *indirect* measurement procedure the analysis of properties of the dielectric as polarisation currents and frequency domain spectroscopy were investigated. Measurements in time and frequency domain determined the influences of moisture, temperature, aging, oil impregnation and pressboard type on the dielectric properties. Based on these measurements new software to analyse dielectric properties of real power transformers was written.

Finally the methods were applied to twelve transformers for on-site moisture evaluation comparing conventional to new approaches. Equilibrium diagrams in the shape of moisture isotherms in all cases delivered credible results. Within this work the developed dielectric analysis software proved its capability to compensate for conductive aging by-products and provide dependable results for moisture assessment of power transformers.

Keywords

Power transformers, condition assessment, oil-paper-insulation, moisture content, Karl Fischer titration, capacitive probes, adsorption, solubility, moisture equilibrium, isotherms, relative saturation, dielectric response measurements, polarisation and depolarisation currents, frequency domain spectroscopy

1 Introduction

"The dryness and ageing state of the oil-paper insulation is a key factor in both the short and long term reliability of a power transformer." [Gubanski 2004]

1.1 Power Transformers in a Liberalized Energy Market

Power transformers are the most expensive links in the chain of transmission network for electrical energy connecting generation to utilization. Nowadays three factors stress transformers: the increased demand for electrical energy, whereas the average age of transformers increases as well and maintenance strategies change forced by the cost pressure in liberalized energy markets. Electric utilities try to suspend the investment in new devices and shift maintenance from time based to condition based strategies. To realize this strategy the demand for new diagnostic methods arises, methods which reliably evaluate the actual condition of the equipment.

Among aging phenomena, moisture in the liquid and solid insulation results into a frequently discussed problem since it decreases the dielectric withstand strength, accelerates cellulose decomposition and causes the emission of bubbles at high temperatures. This work introduces moisture measurement methods, which provide dependable results for testing and maintaining oil-paper-insulated power transformers.

Consequences of Liberalisation

Deregulation and liberalisation of the energy market causes three effects: (1) new evaluation criteria for safety of energy supply, (2) changed energy flows with transmission over long distances and (3) increased use of renewable energy sources. Power utilities try to decrease service costs in order to increase benefits. To achieve this goal, the utilities are forced to fully utilize the capacity and lifetime of equipments, reduce costs of specialized stuff and revise maintenance strategies.

Transformer Aging as a Worldwide Issue

In all developed countries large proportions of the transformer fleets are approaching the end of their design life. For example in the United States the average age of large electrical transformers is 35 years; the design life is 40 to 50 years [TD-World 2007]. The authors conclude: "The data we see indicates that of the 110'000+ large electrical transformers installed in the United States, up to 2 % will fail this year; that is 2200 transformers. ... Utilities need quality,

reliable, maintenance free solutions that provide timely information on the transformers health." Economical interests boost this statement and it mirrors a worldwide trend.

In Germany, a recent investigation found the average age of transformers for 110 kV rated voltage is 31 years, for 220 kV 34 years and for 380 kV 30 years [Gernandt 2006]. The authors word the demand to precisely know the error rate of the equipment and appropriate diagnostic procedures to evaluate these components.

Failures and Aging of Power Transformers

Breakdowns cause not only economical losses, but slur the utilities reputation as well. With increasing age and declining withstand strength the failure probability increases. [Gernandt 2006] published failure statistics of transformers in the German grid and related them to the operating time. Figure 1 illustrates failures of power transformers classified by voltage level and inducing subsystem and the dependence on operating time. The statistic involves transformers disconnected because of a breakdown or a failure signal, resulting in a relative failure frequency of 0,013 failures / year. The small number of old transformers increases the 90 % confidence interval at longer operating times. Moisture affects the failure rate of the active component, partly that of bushings and may contribute to unidentified failures in Figure 1.



Figure 1: Failures at power transformers classified by voltage level and location (left) and as a function of operating time (right). For the relative failure frequency a 90 %-confidence interval is depicted as well.

Test and Maintenance Strategies

Power transformers are electrically tested and maintained by various strategies facing different goals. Factory tests are followed by start-up tests on-site before commissioning. Both tests are to confirm a span of life for the apparatus of

about 30 years. The later performed maintenance tests are to reduce unplanned shutdowns, increase safety and also to avoid personal injuries.

Maintenance strategies shift to more sophisticated approaches:

- *Corrective maintenance* is a reactive mode only initiated by faults, break downs or repairs.
- *Time based maintenance* consists of periodic actions advised by manufacturers recommendations, industrial standards and own experiences.
- *Condition based maintenance* minimizes costs compared to the time based approach, which may involve needless work.
- *Reliability centred maintenance* considers the probability of a failure in a forward-thinking approach.
- *Risk based maintenance* takes both the likelihood and consequence into account and prioritizes equipment and their sub-components accordingly.

The more advanced methods especially require dependable knowledge of the transformer system, provided by reliable diagnostic methods.

An Overview of Diagnostic Methods for Power Transformers

Condition assessment and diagnostics at power transformers may be organized basic diagnostics. advanced diagnostics. on-line monitoring and as investigations during repair or post mortem. Commonly the transformer operator performs basic diagnostics such as oil analysis and simple electric tests on a periodical routine, see Table 1. If these tests indicate deviations from original conditions or exceeded limits, advanced or specialized methods are applied. These methods are suited to the detected or assumed failure and mostly require disconnection of the transformer. On-line monitoring provides a continuous assessment of the actual transformer condition but cost restrictions limit this method to important or new, large transformers.

During repair or post mortem investigations sampling of paper and pressboard becomes possible. This is the only way to directly assess moisture in the solid insulation and the degree of polymerisation. This thesis will provide two indirect methods to assess moisture in the solid insulation.

Technical and economical aspects determine subsequent actions after condition assessment. Technical aspects consist of the exterior condition, operations of all subsystems, condition analysis of the active component and the service history. Economical aspects are the remaining life time, risk calculations, follow-up costs of a breakdown, electrical losses inside an aged transformer and the future development of the electrical grid [Leibfried 2002].

Sequence	Parameter or diagnostic method					
Basic diagnostic	Exterior condition					
(periodical routine)	Oil analysis	- Breakdown voltage				
	(on-site sampling)	- Water content				
		- Neutralization number				
		- Colour				
		- Dissipation factor				
		- Dissolved gas analysis DGA				
	Electrical measurements	- Winding resistance				
	(offline)	- Insulation resistance				
		- No load current				
		- Capacitance and dissipation factor				
Advanced diagnostic	- Transmission ratio					
(offline, after	- Short circuit impedance					
indication)	- Dielectric response methods (moisture in pressboard / paper					
	- Frequency range analy	rsis FRA				
	- Partial discharge meas	urements				
	- Furanes in oil (on-site	sampling)				
On-line monitoring	- Voltage, current					
(important units)	- Oil temperature, pressure, hydrogen content, moisture of					
	- Tap changer position, load current, motor current					
	- Cooling unit, ambient	temperature				
	- Bushing capacitance					
	 Partial discharges 					
During repair /	- Degree of polymerisat	ion				
post mortem	- Moisture in pressboard	l / paper				

Table 1: Diagnostic methods applied on power transformers

1.2 Dangerous Effects of Moisture

Moisture in oil-paper-insulations causes three dangerous effects: it decreases the dielectric withstand strength, raises the emission of water vapour bubbles and accelerates cellulose aging.

Decrease of Dielectric Withstand Strength

Moisture decreases the dielectric withstand strength which is the most important property of oil apart from heat conduction. Breakdown processes start in areas of low density, the so called micro bubbles. Theories assume, that current injection generates these bubbles especially at the negative electrode [Butcher 2005]. Moisture, aging by-products, particles and oil pressure influence the breakdown voltage.



Figure 2: Breakdown voltage in new insulation oil Nynas Nytro 3000X depending on moisture relative to saturation and acidity (Total Acid Number TAN). Spherical electrodes with a gap of 2,5 mm were used according to IEC 156/1995-05.

Figure 2 illustrates the influence of moisture relative to saturation and acidity on the dielectric withstand strength of oil [Koch 2007.1]. Water molecules dissociate themselves and enable acids to dissociate supporting breakdown processes with charge carriers. The relative availability of water molecules, reflected by the moisture content relative to saturation, is responsible for influencing breakdown processes. Above saturation level water is not longer dissolved, but forms a dispersion of small droplets without any additional influence on breakdown voltage [Kuechler 1996]. Moisture relative to saturation also includes the influence of temperature.

Thus the moisture content relative to saturation [%] is more appropriate to describing the influence of moisture on breakdown processes than moisture relative to weight [ppm]. Unfortunately standards like IEC 60422 give moisture limits for transformer oils relative to weight (ppm as parts per million), disregarding the influence of temperature and aging.

Evolution of Water Vapour Bubbles

One of the issues of wet transformers is the emission of gaseous bubbles from wet insulation paper at high temperatures. This phenomenon results in two dangerous effects:

- Formation of vapour-filled cavities (bubbles) on the insulation surface and in oil with dramatic decrease of the dielectric strength
- De-impregnation of the turns insulation and displacement of insulation oil

Insulating liquids derive the dielectric strength from their much higher density compared to gases. The breakdown process starts with a microscopic area of large distances between corpuscles, where ions or electrons can initiate avalanches. These microscopic bubbles originate from current impulses on an electrode. The next current impulse injects charge carriers into the bubble, leading to current amplification and finally to breakdown [Butcher 2005]. When



wet oil-paper-insulation is heated the bubbles exist already before application of an electrical field, thus the dielectric withstand strength decreases significantly.

Figure 3: Bubble inception temperature depending on moisture content for new Kraft paper, thermally upgraded paper and thermally degraded Kraft paper in new Shell Diala D (left) and for new Kraft paper in service-aged oil, thermally degraded Kraft paper in service-aged oil[Koch 2007.2] in comparison to the results from [Oommen 2001](right)

The diagram to the left of Figure 3 shows the bubble inception temperature in new oil Shell Diala D with new Kraft paper, thermally upgraded paper and thermally degraded (aged) Kraft paper [Koch 2007.2]. New Kraft paper has the lowest inception temperature. For thermally upgraded paper the bubble inception starts at higher temperatures because of its microstructure. Filler materials make it resistant to thermal aging and decrease the volume of pores and capillaries. The capillary force increases and bubbles will be driven out at higher temperatures.

Surprisingly for thermally degraded Kraft paper the inception temperature increases. During thermal aging the active parts of the cellulose chains (OH-groups) become attracted to each other. The number, diameter and volume of pores diminish and the capillary forces increase [Weigert 1999].

Aging of oil besides paper aging is of crucial importance for the inception temperature. Aging by-products decrease the surface tension and therefore the bubble inception temperature. The diagram to the right of Figure 3 illustrates this effect for a service-aged transformer oil Shell Diala K 6 SX from 1965 with a total acid number of 0,5 mg KOH/g oil.

Accelerated Aging of Cellulose

Cellulose in power transformers deteriorates through oxidation, hydrolysis and pyrolysis, resulting in a decreased mechanical withstand strength. Hydrolysis is

considered to be the dominating aging process, since this reaction takes place even at service temperatures (activation energy 37 kJ/mol at 4 % moisture content to 86 kJ/mol at 1 % moisture, [Hoehlein 2004]). At hydrolysis the long cellulose chains break down into smaller pieces. Water is added in this reaction, acids serve as a catalyst [Lundgaard 2004]. Thus the degree of polymerisation (DP, number of jointed glucose rings per cellulose chain) decreases from 1000-1500 to 200-400 at the end of the cellulose life span. Figure 4 illustrates the life expectancy of paper, supposing a starting DP of 1000 and ending DP of 200. A temperature increase of approximately 8°C halves the life expectance and moisture decreases it further.



Figure 4: Expected life for solid insulation depending on moisture content and temperature [Lundgaard 2004]

1.3 Sources of Water Contamination

Water contamination of power transformers is caused by three sources: residual moisture after manufacture, ingress from the atmosphere and aging of cellulose and oil. A new transformer leaves the factory having 0,4-1 % moisture by weight in the cellulosic insulation. The overall water contamination during service of around 0,1 % per year results in a typical value of 3 % for a 30 years old transformer (section 0).

Residual moisture remains in the bulky insulation components, which require a long drying time in the manufacturing process. During the temperature cycling of the transformer service this moisture migrates into the thinner insulation structures with oil as a moisture carrier.

The atmosphere is considered to be the main source of moisture [Sokolov 2008] and three mechanisms contribute: Firstly air humidity becomes adsorbed during direct exposure to air which happens at installation and repair. Secondly moisture can ingress into the tank due to the difference in water vapour pressure

in the atmosphere and the transformer gas space or oil. Thirdly for temperature drop of open breathing transformers the atmospheric pressure is higher than the pressure inside the tank. Thus air ingresses and also moisture, if the dehydrating breather cannot completely dry the air. However, the contamination rate because of breathing strictly depends on the operation mode and breather condition of the specific transformer [Koch 2008]. Additional water comes through typical leaks as the top seal of draw-lead bushings, the seals in explosion vents and leaks in forced-oil circulation systems between the main tank and the coolers. At oil pumps air can be sucked in because of the "negative" pressure created before the pumps.

Aging of cellulose materials leads to molecular chain scissions and the formation of by-products such as water and furanic compounds. The average water production per chain scission seems to be around 0,2 % by weight, unfortunately this result varies between different research groups. The aging model proposed by [Sokolov 2003] indicates oxidation, pyrolysis and hydrolysis of cellulose and oxidation of oil as reactions which contribute to water contamination. However, hydrolysis also consumes water. Further research is needed to better quantify the total amount of water generated by aging of paper and pressboard. Figure 5 illustrates the sources of moisture contamination for transformers in service.



Figure 5: Sources of water contamination in power transformers

According to [Sokolov 2008] the hot winding paper generates water due to aging. From the hot areas water migrates through the oil to the colder thin

structures of the main insulation between high and low voltage winding, which thus accumulate moisture (see also p. 59).

1.4 Aim and Structure of this Work

Although moisture in oil-paper-insulations causes various detrimental effects and knowledge about the moisture content is of prime importance for the operators, the commercially available measurement techniques often provide conflicting results. Figure 6 compares moisture analysis results on the same transformer obtained using four different methods: two kinds of dielectric response methods (recovery voltage method RVM, frequency domain spectroscopy FDS) and two different equilibrium diagrams [Oommen 1983], [IEEE 1995]. One of the depicted results suggests that the insulation is very wet with more than 4 % water in cellulose, where as the others show less water, culminating in only 0,5 % for the FDS method, which would imply a very dry insulation.

The depicted findings make a dependable moisture evaluation for subsequent actions like drying impossible. This thesis will enable the reader to judge even such contradictory results, improve the existing techniques and introducing a new method to reliably assess the moisture content in the solid insulation.





The present research work started in 2003 within in the European Union sponsored project REDIATOOL¹, involving three universities, the German power utility RWE and three international transmission system operators. Also two German power transformer manufacturers collaborated in this work; Areva Schorch Transformatoren GmbH in Moenchengladbach and Siemens AG, PTD

¹ European research project REDIATOOL: "*Reliable Diagnostics of HV Transformer Insulation for Safety Assurance of Power Transmission System*", contract NNE5/2001/472

Transformatoren in Nuremberg. The research at first focused on dielectric response methods, but later involved equilibrium methods for the measurement of water in transformers.

Objectives of this Thesis

The objectives of this thesis are to:

- enhance knowledge about water sorption, diffusion and equilibrium especially under aged conditions,
- introduce a new approach to describe and measure moisture in power transformers by means of the relative saturation in oil and cellulose,
- explain and eliminate errors of conventional moisture measurement methods, e.g. enlighten difficulties with Karl Fischer titration and equilibrium methods,
- improve interpretation schemes for dielectric response methods, e.g. by considering the properties of oil conductivity and conductive aging by-products,
- provide a reliable data pool for the analysis of on-site measurements,
- enable non-experts to achieve reproducible results over time,
- give good correlation of new approaches to other methods, e.g. by comparison of on- and offsite measurements,
- and finally to provide a moisture evaluation, that enables decisions to be made about further actions like drying. An accuracy of ± 15 % suffices for a classification into the stages dry, fair and wet.

Structure

The chapters 2, 3 and 4 introduce the particular topic with a literature review, describing the current state of the art.

Chapter 2 investigates direct moisture measurement techniques of oil and cellulose using Karl Fischer titration and capacitive probes. It aims to provide dependable assessments by direct techniques before new indirect techniques are developed in the following chapters.

Chapter 3 discusses moisture measurements based on equilibrium conditions. Beside the common mineral oils, vegetable oils are also covered. Starting from the crucial weaknesses of conventional equilibrium diagrams improvement steps are introduced, resulting in a new approach to measure and use moisture saturation to describe moisture in power transformers. Chapter 4 introduces dielectric diagnostic methods and deals with approaches to improve their reliability, e.g. measurements of the dielectric properties at new and aged pressboard samples, properties of oil conductivity, consideration of conductive aging by-products and influence of pressboard type. It closes with a description of new software to analyse dielectric measurements.

Chapter 5 confirms the new approaches using examples of on-site measurements on real power transformers, performed by comparing results of different measurement methods. It also quantifies the influence of moisture migration on on-site dielectric measurements.

Chapter 6 finally summarizes the main findings of the research work and suggests topics of further research.

2 Direct Measurement in Oil and Cellulose

"There is a need to check/verify the estimates of water content determined by the dielectric response techniques by comparison with basic chemical measurements, although it is accepted that this will introduce other uncertainties." [Guuinic 2001]

2.1 State of the Art

Before assessing water in transformers with new and indirect approaches, it is necessary to achieve reliable results with conventional and direct measurement methods on oil and cellulose. These direct measurements then serve as a standard for the new and indirect approaches. Improvements of conventional methods are not the main objective of this thesis, although there is an obvious need.

The titration according to Karl Fischer represents the state of the art method to measure the water mass in the solid and liquid insulation. Capacitive probes are often used to measure moisture relative to saturation, especially under on-site and on-line conditions.

This chapter will introduce both measurement principles and special care for the reliability and accuracy of the methods, since they are the reference for the later introduction of new indirect methods.

A Round Robin Test on Karl Fischer Titration

To evaluate the moisture content in the liquid and solid insulation the titration according to Karl Fischer is not only widely used, but also serves as a benchmark for other methods such as dielectric response methods. Actually it is affected by several influences:

- There is always moisture ingress from the atmosphere during sampling, transportation and sample preparation.
- Cellulose binds water with chemical bonds of different strengths. It is uncertain whether the thermal energy releases all the water. Heating temperature and time certainly changes the released water.
- Individual laboratories treat differently constraints that are not covered by standards, such as the solvent for oil extraction.

• Sometimes direct injection and heating method lead to different results for moisture in oil. This might be due to the influence of oil additives and aging by products.

For these reasons round robin tests revealed an unsatisfactory comparability of different laboratories. The following paragraphs report the conditions and results of one of them.

Laboratories and Samples

Eight laboratories from four European countries took part in a round robin test. The laboratories measured the water content in paper relative to weight and the water content in oil relative to weight following their standard procedure. Table 2 describes the equipment used in the test.

Lab	Instrument for Paper samples	Heating temp.	Instrument for Oil samples
А	Coulometric, heating with oven	160°C	Coulometric, direct injection
В	Coulometric, heating with oven	200°C	Coulometric, direct injection
С	Coulometric, heating with oven	135°C	Coulometric, heating with
			oven
D	Coulometric, heating with oven	135°C	
Е	Volumetric	-	Volumetric
F	Coulometric, heating with oven	140°C	Coulometric, heating with
			oven at 80°C
G	Coulometric, heating with oven		Coulometric, direct injection

Table 2: Titration techniques used in the round robin test

Three oil and paper samples filled flasks made of polyethylene and were the measurement objects. Two of the samples consisted of new Kraft paper and new insulation oil Nynas Nytro 3000 having a relative moisture saturation in oil and paper of 2,7 % resp. 4,7 %, both measured at 80°C. The third sample was a service-aged paper from 1954 in service-aged Shell K 6 SX having a relative moisture saturation of 33 % at 27°C. This oils total acid number was very high with 0,49 mg KOH / g Oil.



Measurement Results

Figure 7: Moisture content in paper in % relative to weight as measured by the laboratories (left) and deviation of each laboratory from the average (right)

The results of moisture in paper as depicted in Figure 7 show a reasonable conformance. One systematic influence is obvious: The heating temperature of paper in the oven increases the released mass of water. This is due to chemical bonds of different strengths which keep the water molecules in the paper. Beside this the differences could be explained by stochastic influences only. For the low water content of sample A the comparability is worst and hardly acceptable.



Figure 8: Moisture content in oil in ppm relative to weight as measured by the laboratories (left) and deviation of each laboratory from the average (right)

The results of moisture in oil in ppm (Figure 8) provide a reasonable conformance only for the wet sample C. For the rather dry samples A and B only a trend is recognizable. Systematic differences are obvious. It has to be mentioned, that for dry oils the results also scatter within one single laboratory, a standard deviation of 20 % being not unusual. The following section 2.2 will investigate the influences and give advice on practical problems.

2.2 Karl Fischer Titration

2.2.1 The Karl Fischer Titration Technique

Karl Fischer titration is a method in analytical chemistry that determines trace amounts of water in a sample using volumetric or coulometric titration. The German chemist Karl Fischer (1901-1958) introduced this titration method in 1935 [Fischer 1935]. Titration basically means to add a reagent of known concentration (titre) to an unknown substance until the concentrations are balanced. For volumetric titration the added volume is measured and the required value (here water content) is calculated by a stoichiometric equation. The sensitivity of volumetric titration is limited to some 10 μ g water so that it is hardly applicable for dry insulation oils. With coulometric titration the reagent iodine is generated by a generator electrode and the need of its standardization is eliminated. (1) is a simplified stoichiometric equation illustrating the reactions taking place in the titration vessel. The detection limit of coulometric titration reaches down to several μ g of water.

$$2H_2O + SO_2 + I_2 \rightarrow H_2SO_4 + 2HI \tag{1}$$

Equation (2) then calculates moisture relative to weight W as the ratio of the measured water mass to the mass of dry and oil free paper m_d multiplied with 100 %. The water content W can also be related to the wet mass of the sample, but in this investigation the first form is used. Moisture ingress into the titration system as defined by the drift D_t (µg/min) and the humidity in an empty sample vessel (blind value $m_{H2O,B}$ in µg) must be subtracted from the total moisture income m_{H2O} .

$$W = \frac{m_{H_2O} - t \cdot D_t - m_{H_2O,B}}{m_d} \cdot 100\%$$
(2)

The moisture of the material could be brought into the electrolysis cell (Figure 9) by three methods:

1. It is possible to inject the material directly into the reaction vessel, e.g. oil through a syringe. This method pollutes the reagent and side reactions of ageing products or inhibitors with the reagent are possible.

- 2. The water from a cellulose sample can be extracted by immersion into a hydrophilic medium, e.g. methanol. Methanol and water will then be injected into the reaction vessel. Because of the slow extraction this methanol extraction is nowadays rarely applied.
- 3. The sample can be heated in an external oven and a gas stream with water vapour is lead into the reaction vessel. Advantageously the reaction vessel remains clean of pollutants and also of ageing by-products, depending on their boiling point. A drawback of this method is the blank value of the gas stream and of the heating vessel. This decreases the sensitivity compared to the direct injection method, especially for oil. Figure 9 (right) illustrates a Karl Fischer titration of a pressboard sample using an external oven.



Figure 9: Electrolysis cell for coulometric Karl Fischer titration (left) and currently measured and accumulated water mass at a typical water titration in pressboard with external heating (right)

2.2.2 Influence of Heating Temperature for Cellulose Samples

Heating with an external oven is widely used for water in cellulose measurements. Cellulose binds water with chemical bonds of different strengths. Water measurements should reveal all water from the material; however it is uncertain whether the thermal energy does release all the water. Heating temperature and time strictly change the amount of released water molecules. According to IEC 60814 the temperature should be 130-140°C. [Kleboth-Lugova 2004] found, that for pressboard samples only a temperature of 180°C leads to results comparable with the methanol extraction method. On the other hand the upper limit for the heating temperature is given by pyrolysis or oxidation of cellulose with subsequent water production in the heating chamber. This occurs for paper above 170°C and for pressboard above 200°C.

Several measurements at various increasing temperatures determine the heating temperature for the material to be measured. Here the optimal heating temperature was found to be 160°C for insulation paper and 180°C for pressboard. These temperatures not only ensure that more water is detected, but that the measurement is faster (e.g. 5 min at 170°C instead of 12 min at 120°C) and the single measurements are less scattered. Figure 10 illustrates the influence of heating temperature on water content and scattering.

2.2.3 Solvent for Oil Extraction for Cellulose Samples

After titration it is necessary to extract oil from paper in order to obtain the true dry weight of the paper sample for application in equation (2). Unfortunately the standard IEC 60814 only states an "appropriate chlorine-free solvent". Some laboratories do not separate oil from paper and thus obtain too high a paper mass. Others use methanol as oil solvent. Indeed methanol cannot dissolve oil, a dispersion arises instead of a solution so a lot of oil remains in the paper. For this reason heptane was investigated in comparison to methanol. Its nonpolar molecule structure enables heptane to extract the likewise nonpolar mineral oil from cellulose. Other paraffin mixtures with boiling points of 70-90°C are applicable as well (e.g hexane).



Figure 10 shows the improved qualification of heptane in comparison to methanol. Heptane releases more oil from paper, the dry weight of paper decreases, thus according to equation (2) the obtained water content increases. Here a titration system called Aqua 40.00 by ECH Germany with head space module Coulomat Oven AG from Riedel de Haen as reagent was used.

2.2.4 Influence of Aging By-Products on Oil Titrations

Aging by-products from oil degradation and inhibitors may influence the Karl Fischer reaction of oil samples with direct injection. Aging of oil causes

alcohols, aldehydes, ketones, acids and sludge. It is assumed, that aldehydes and ketones react with methanol of the reagent to acetales and ketales with water as a byproduct. To confirm this suspicion, a coulometric titration cell with direct injection titrated various oil samples (new, aged, inhibited). Two different reagents from Riedel de Haen were used; Hydranal Coulomat AG with methanol and Coulomat AK without, which is especially suitable for titrations in aldehydes and ketones preventing side reactions.

A comparison of the reagent Coulomat AG to Coulomat AK, which is not sensitive to ketones, was published by [Koch 2007.3]. The results scatter, but the reagent AG systematically measures 10 % more water compared to AK, which might be explained by side reactions with ketones. Still one has to consider that 10 % is a minor deviation compared to that in the round robin test in Figure 8. It is not advisable to always use reagents that are insensitive to ketones and aldehydes, since their water absorption capacity is smaller.

2.2.5 Direct Injection versus Heating Method for Oil Titrations

Three variations of the Karl Fischer technique for oil titration were compared:

- volumetric titration using a Schott Titroline Alpha, with reagent Coulomat Solvent Oil
- coulometric titration with direct injection using an ECH Aqua 40.00 and Coulomat Oil by Riedel de Haen as reagent
- coulometric titration with heating chamber for oils using an ECH Aqua 40.00 and Coulomat Oven AG by Riedel de Haen as reagent, heating temperature at 80°C and oil volume 10 ml

Of course all the techniques should result in the same moisture content in oil.

One remark about water in oil measurements by heating in an external oven: Transformer oils usually have very low water contents of sometimes less than $5 \mu g/g$ oil. Unfortunately for the heating method the water mass should exceed 50 μg to be distinguishable from the drift and the blank value. Therefore an oil volume of at least 10 ml should be heated. It is not sufficient to use the vials from paper measurements since their useable volume is 2,5 ml only, so a special heating chamber for oil is necessary.

Figure 11 depicts the results of the comparative measurements at various new and aged oil samples. For new oils all the three methods give comparable results, although also here a deviation of 20 % appears. Volumetric titration indicates the highest water contents, followed by direct coulometric titration.

Coulometric titration with an external heating chamber results in the lowest water content.



Figure 11: Moisture in various oils for volumetric titration, coulometric titration with direct injection and coulometric titration with external heating

In aged oils the situation becomes even worse. Here volumetric titration measures the highest water contents. Unfortunately the coulometric titration with external heating in some oils was not able to measure their water content. The drift did not decrease to the original value but remained at 20-30 μ g/min, which possibly indicates a side reaction. This leads to water contents of some 100 ppm as in the round robin test in Figure 8. As a conclusion, the external heating method cannot be recommended for aged oils, while aged materials are the common case in transformer diagnostics.

The inferences from the investigations of section 2.2 resulted in a guide for reliable Karl Fischer titrations, which can be found in appendix 7.1 of this thesis.

2.3 Capacitive Probes

Capacitive probes indicate moisture relative to saturation. They consist of two electrodes with a hydroscopic dielectric, e.g. a polymer, see Figure 12. Water molecules penetrate into the polymer depending on the relative saturation of the ambient material. Water penetration *c* then changes the capacitance *C* because of the high relative permittivity of water $\varepsilon_r = 80$. An RC-oscillator measures the

change of capacitance by a change of frequency, this variance is calibrated to relative saturation.



Figure 12: Schematic drawing of a polymer thin film probe (left) and water in the dielectric attached to polyimide molecules by hydrogen bonds (right)

$$C = \frac{\varepsilon_0 \cdot \varepsilon_r \cdot A}{d} \quad \text{and} \quad \varepsilon_r = \varepsilon_i + \varepsilon_w \cdot c \tag{3}$$

Advantages of capacitive probes are very easy application even on-site and online, continuous measurements, fast response, good stability and repeatability, no hysteresis, broad humidity and temperature range, low temperature coefficient and high accuracy compared to sampling and transportation to laboratory with subsequent Karl Fischer Titration.

Accuracy of Capacitive Probes

A comprehensive compensation for temperature effects and nonlinearities is necessary, especially for the low humidity range. Beyond this molecules other than water may likewise diffuse into the polymer and thus change the capacitance, leading to a measurement error [Wiederhold 1997]. This influence is considered to be small, since e.g. aging by-products have a low permittivity compared to water.

Strong acids may corrode the electrodes and thus change the capacitance of the arrangement. Therefore a cyclic calibration of capacitive probes is required. Commercially available sensors differ in their long term stability under stress and the time to achieve a stable reading.

In dry, not flowing oil capacitive probes often need a very long time to come into equilibrium with the oil in order to show a stable reading. Here only a small number of water molecules diffuse into the polymer, causing only a slow change of capacitance. In a flowing oil the equilibration process occurs much faster generally in less than five minutes.

Calibration of Capacitive Probes

Capacitive probes can be calibrated using saturated salt solutions in a closed vessel. Saturated salt solutions provide an exactly defined relative humidity in the gaseous phase above them [Greenspan 1977]. Observing temperature, gas circulation and sufficient time the probe is calibrated in such a vessel. Lithium

chloride and sodium chloride calibrated the two probes used here (Vaisala HMP 228) at their typical relative humidity of 12 and 76 %.

There still remains an uncalibrated gap in the range of 0-12 %, which is very important for power transformers. Therefore a comparative measurement with chilled mirror hygrometers was performed in flowing, dry, compressed air. Figure 13 depicts the calibration results; the relative humidity reading as a function of time (left) and for air with varied humidity (right). The self-calibrated capacitive sensors (V1, V2) show an excellent conformance with the chilled mirror hygrometer (CM). In contrast to this an uncalibrated commercially available probe (EE) shows a positive deviation of 4 % relative humidity.



Figure 13 Calibration of relative humidity probes in dry air as a function of time (left) and a comparison of the final end values (right)

Comparability to Karl Fischer Titration

In the last years discussions frequently flared up about the reliability of Karl Fischer titration in comparison with capacitive probes. Some transformer operators listed significant differences between the moisture by weight (ppm) results from Karl Fischer titration and capacitive probes or other instruments based on moisture equilibrium [Altmann 2003]. They concluded that Karl Fischer titration is not valid for aged oils since: Firstly, a deviation between capacitive probes and Karl Fischer titration appears only for acidic oils. Secondly, for aged oils the breakdown voltage was good, although Karl Fischer titration measured a high moisture content. Capacitive probes still indicated a low moisture content.

Answering these arguments, it is at first helpful to look at the market launch of capacitive probes. These probes feature moisture relative to saturation as their original measurement result. As transformer operators were used to moisture by

weight in oil (ppm), moisture relative to saturation (%) was not desired. So the probe manufactures developed arithmetic equations to obtain moisture by weight (ppm). These equations include two constants A and B, that have to be calibrated for every oil, where the probe is applied. Unfortunately this calibration is not usually done. On this basis it is principally incorrect to compare the ppm-result of uncalibrated capacitive probes to the ppm-result of Karl Fischer titration. Karl Fischer titration measures water mass by a chemical reaction while capacitive probes measure moisture equilibrium and therefore the relative moisture content. This explanation applies to all measurement techniques based on moisture equilibrium such as head space methods used in photo-acoustic DGA.

Secondly, the moisture induced breakdown voltage of oil depends directly on the moisture content relative to saturation level, but there is only an indirect relation to moisture by weight in ppm (Figure 2 and [Kuechler 1996]). For new oil a high moisture content *relative to weight (ppm)* means a high moisture saturation, resulting in a low breakdown voltage. The same high ppm-value in an aged oil having a high water solubility results in an unexpectedly high breakdown voltage.

3 Determination by Moisture Equilibrium

"Eureka" – "I have found it!" – Archimedes after discovering volume displacement in water

This chapter describes the indirect assessment of moisture in cellulose via the moisture of the surrounding oil. Therefore one needs knowledge about water solution in mineral oil and adsorption in cellulose, as presented by moisture isotherms. At least a basic understanding of the physical and chemical background of moisture adsorption is also provided. Then moisture diffusion and equilibrium are characterized in terms of thermodynamic equilibrium. The measurement technique of moisture in cellulose by moisture in oil is described. Finally a new description in terms of moisture relative to saturation for water in transformers is favoured above the conventional measure relative to weight.

3.1 State of the Art and Literature Review

Moisture partitioning curves are used in decades to easily examine moisture in paper based on moisture in oil, often, however, with doubtful results (Figure 6). Over the years several scientists have published equilibrium diagrams. Thus it is useful to have an overview of the history of these curves [Du 1999].

History of Moisture Equilibrium Diagrams for Oil and Cellulose

[Fabre 1960] at first reported equilibrium curves for water in oil and Kraft paper. These curves were obtained using direct Karl Fischer measurements in oil and paper. Some years later [Norris 1963] referred to these curves, and they were known as "Norris Curves". In later years the results of Fabre were redrawn by many people in a different format, neglecting the original source.

Another well known set of curves was published by [Oommen 1983]. His results are based on the principle, that the relative humidity is the same in the oil, in cellulose and in air in contact with one another at the same temperature after sufficient time had passed to reach equilibrium (section 3.4). Thus he combined moisture isotherms for oil (moisture in oil versus relative saturation at various temperatures) with isotherms of cellulose (moisture in cellulose versus relative saturation at various temperatures). Oommen used cellulose isotherms of wood pulp as measured by [Jeffries 1960] and his own isotherms. The moisture saturation range below 20 % was not measured but only extrapolated. However, this is the most important range for power transformers.

History of Sorption Isotherms for Cellulose Materials

The research on moisture sorption isotherms for transformer materials, i.e. moisture in cellulose versus relative humidity at different temperatures, reaches

back to 1946. [Piper 1946] combined four groups of data from the literature: data for cotton at high moisture concentrations, data for cotton at low moisture concentrations, data for Kraft paper of low moisture content and data for spruce wood of high moisture content. [Jeffries 1960] compared cellulose to textile polymers and found the water contents of [Piper 1946] to be too high. [Beer 1966] measured vapour adsorption isotherms directly for Kraft paper, thus his data are preferable to the older. [Guidi 1974] also found, that the data of [Piper 1946] derived from cotton was too high. [Oommen 1984] created isotherms based on the data of [Beer 1966]. [Fessler 1987] combined data used by [Piper 1946], published by [Oommen 1984] and own experiments to determine a formula describing water adsorption isotherms for paper. [Griffin 1988] used the formula of [Fessler 1987] for sorption isotherms.

From this review we can see that, although there are many publications there are only a few measurements of transformer materials. These are [Piper 1946], [Fabre 1960], [Beer 1966] and [Oommen 1984]. Many of the data were obtained with extrapolation and combination of other researchers work and are valid for materials used in those times to manufacture transformers. The very detailed investigation of isotherms of various cellulose materials by [Tveit 1966] proves, that it is not sufficient to deal with a "common" cellulose material. Moreover all the equilibrium diagrams disregard aging, so their application often leads to erroneous results as explained in the following paragraph.



Conventional Application of Equilibrium Diagrams

Deriving the moisture content (%) in cellulose from the moisture content in oil (ppm) is a standard procedure for operators of power transformers. This approach consists of three steps: (1) Sampling of oil under service conditions, (2) Measurement of water content by Karl Fischer Titration and (3) Deriving moisture content in paper via equilibrium diagrams (e.g. by [Fabre 1960], [Du 1999] and [Oommen 1984], shown in Figure 14) from moisture in oil.

Unfortunately this procedure is affected by crucial errors:

- Sampling, transportation to the laboratory and moisture measurement by Karl Fischer titration,
- Equilibrium conditions are rarely achieved (depending on temperature after hours/days/months),
- A steep gradient and high uncertainty in the low moisture region compounds the accuracy,
- One obtains different results using different diagrams from the literature,
- A vertical moisture distribution in transformer windings exists because of the temperature gradient of up to 30 K in naturally cooled transformers,
- Equilibrium depends on moisture solubility in oil and moisture adsorption capacity of cellulose.



The validity of equilibrium diagrams is restricted to the original materials that were used to establish the diagrams. Aging especially changes the moisture adsorption capacity substantially. Figure 15 displays the graphs for moisture equilibrium of new Kraft paper with new oil at 20, 40, 60 and 80°C. Additionally for 60°C it shows moisture equilibrium for new pressboard in new oil and for aged Kraft paper and aged pressboard in aged oil. Assuming the moisture content in oil is 20 ppm these curves lead to a moisture content in new paper of 2,9 %, in new pressboard of 2,6 %, in aged paper and aged oil it is 2,1 % and for aged pressboard and aged oil 1,5 %. Thus equilibrium diagrams

not adapted to the material and its aging state are inapplicable to calculate moisture in paper from moisture in oil.

[Oommen 2003] already emphasized the uncertainties using equilibrium diagrams, but underestimated the influence of oil aging. He stated "... since the paper also ages ... the equilibrium curves are not significantly affected." In reality with aging the water solubility in oil increases by several hundred percent, while that of paper decreases only by around 50 %. Because of the underestimation of aging by-products other research groups also achieved wrong results as illustrated in Figure 6.

Need for New Equilibrium Curves and Interpretation Schemes

The present equilibrium curves suffer from various drawbacks: There is only a small number of real measurements, they do not regard new transformer materials and aging is basically underestimated. Thus the following paragraphs depict novel moisture isotherms measured at various materials and also various equilibrium curves. Proper ways to avoid errors during the measurement are described as well.

Even more important is the introduction of the relative moisture saturation for transformer materials, which is not only easy to measure but also gives more information about the detrimental effects of water.

3.2 Adsorption of Water in Cellulose Materials

3.2.1 Moisture Sorption Isotherms

Moisture sorption isotherms relate moisture relative to weight W of a material to the relative humidity RH of the surrounding air under equilibrium conditions at a specific temperature. The shape of sorption isotherms characterises the inner structure and the affinity for water of a material. Moisture sorption isotherms in ideal nonpolar liquids are straight lines (Figure 24). For solid materials one can discriminate between sorption at the surface (minerals) and sorption into the material (porous materials and also plastics).



Figure 16: Types of moisture sorption isotherms for solids [Brunauer 1940]. Type II characterises porous materials based on wood.
[Brunauer 1940] classified sorption isotherms as depicted in Figure 16. Type I represents a mono layer adsorption at the surface. The S-shaped type II is typical for porous media like paper, pressboard and wood. Low water contents form mono-layers while at higher contents the water molecules are attracted to multiple layers. The curve shape at 100 % relative humidity shows an asymptotic behaviour. The reason is, that at 100 % RH the pores contain an unquantifiable amount of free water. Whether the amount of free water is e.g. 3 % by weight or 10 % by weight, in both cases the induced water vapour pressure will come up to 100 % relative humidity in air. Type III isotherms are typical for plastics, which are hydrophobic for low water contents but water swellable at high water contents. Type IV and V follow from II and III, if the last water film closes the pores, disabling the development of free water.

Cellulose Structure

The micromolecular structure determines the mechanical and chemical properties of cellulose. Cellulose materials consist of wood fibres and therefore contain cellulose (75-85%), polyose (10-20%) and lignin (2-6%). The percentage composition varies with wood species and location and also plays an important role in water adsorption.

Cellulose is a macro molecule made by glucose rings that are linked by glucosidic bonds (Figure 17). The number of glucose rings with 1000-1500 for new materials is the degree of polymerisation and sets the mechanical strength of the material. At a degree of polymerisation of 200 the material perishes and has lost its mechanical withstand strength, the end of the insulations life span is reached. During aging the chemical processes hydrolysis, pyrolysis and oxidation cause chain scissions and produce aging by-products as for example lower molecular acids [Sokolov 2003], [Lundgaard 2004].



The cellulose molecules are bound by hydrogen bonds to crystalline elementary fibrils. These fibrils form micro fibrils and micro fibrils are connected to macro fibrils or fibres, visible with an optical microscope. Various models, reported by [Weigert 1999], describe the distribution of the polyose-lignin-gel within the elementary cellulose fibrils.

Water in Cellulose

Water adsorption takes place in amorphous areas of the cellulose, in the polyose-lignin-gel and in the pores of the cell walls. Water swells the amorphous structures and increases the fibre flexibility, [Weigert 1999].

Because of its polar molecular structure cellulose can attract large quantities of similar polar water molecules. Water exists in the following gradations [Weigert 1999]:

• Monolayer adsorption

At low water contents the water molecules are strongly bound to the OHgroups of the glucose chains by hydrogen bonds, forming a monolayer (Figure 18). At a hydrogen bond two atoms with a high electronegativity bind one hydrogen atom (proton) in between. This water is non freezing bound water.

• Polylayer adsorption

At higher water contents weaker Van der Vaals bonds attract the water molecules forming multiple layers. The weak Van der Waals forces result from electrostatic interactions between molecules and dipoles. Now the cellulose begins to swell.

• Free water

At water contents near the saturation the water fills pores and interstices, held by capillary forces. Water can here form the usual water clusters and it freezes at low temperatures.



Figure 18: Adsorption of water molecules in microcapillaries of cellulose materials

Figure 19 shows the S-shaped isotherms of cellulose materials with three characteristic parts. An initial rapid rise of water content indicates the formation of a "monolayer" of water in the cellulose structure. Part two at further increased vapour pressure shows a less steep increase in moisture content associated with "polylayer" adsorption. An unquantifiable amount of free water in pores causes the third part at high moisture contents.

The adsorption process does not require activation energy, thus the corresponding isotherm lies below the desorption isotherm. For desorption, energy is required to loose the bond. At increasing temperature or pressures the water adsorption capacity decreases, thus the isotherm shifts to the right.



3.2.2 Measurement of Isotherms at Cellulose Materials

Investigated Materials and Measurement Setup

The cellulose types explored for isotherms were pressboard, Kraft insulation paper and thermally upgraded paper. Thermally upgraded paper is a normal Kraft paper stabilized for thermal aging, containing some additives which e.g. block the chemically active OH-groups. To obtain aged materials some new cellulose samples were thermally degraded by heating at 130°C under wet air for one week resulting in a distinctly decreased mechanical strength. Additionally a service-aged pressboard from a transformer manufactured in 1954 was investigated. Its corresponding oil had a total acid number of 0,49 g KOH / g oil, which is very high.

The sorption isotherms were measured at the temperature and moisture range usual for power transformers, that is 20-80°C and 0,4-6% moisture by weight. The graphs are superpositions of several measurements, no discrimination between adsorption and desorption was made. About 100 g of each cellulose material was filled in separate polyethylene flasks. Two capacitive probes (Vaisala HMP 228) as described in section 2.3 measured the relative humidity of the air in the flasks after waiting for equilibrium with the investigated cellulose materials. Coulometric Karl Fischer titration with a heating oven (ECH Aqua 40.00) determined the moisture by weight applying the procedure according to section 7.1.

Moisture Isotherms for Cellulose Materials

Figure 20 to Figure 22 show moisture adsorption isotherms, i.e. moisture relative to weight as a function of moisture relative to saturation for the temperatures 23, 40, 60 and 80°C.



Figure 20: Isotherms for new pressboard (left) and new Kraft paper at 21-80°C



*Figure 21: Isotherms for thermally degraded pressboard (left) and Kraft paper at 23-*80°C

The isotherm shape agrees well with the theory of [Brunauer 1940]. The measured cellulose materials differ in their moisture sorption capacity. The steeper the slope of the curves, the higher is the moisture sorption capacity. New Kraft paper adsorbs the most water molecules, followed by new pressboard and new thermally upgraded paper. It must be admitted, that this difference is not necessarily a material property but might result from different base materials (wood) and its ratio of cellulose to the polyose-lignin-gel.



Figure 22: Isotherms for new thermally upgraded paper (left) and for service aged pressboard at 21-80°C

The isotherm of the service-aged pressboard from 1954 indicates the lowest water adsorption capacity (Figure 22). This is caused by real aging conditions and also by the material used at that time.

Aging seems to decrease the moisture adsorption capacity (Figure 21 and Figure 22). This agrees well with findings of other research groups, e.g. [Brandt 1989], [Shayegani 2007]. A contingent explanation gives an effect called hornification, well known at the paper industry. Here during thermal aging the OH-groups of the cellulose chains become attracted to one another, less able to attract water molecules. These materials become rigid and are less water swellable. Beside this, acids might become attracted to the OH-groups making them less active in attracting water.

Accuracy of the Results

The accuracy of the isotherms depends on the accuracy of each measurement system, here capacitive probes and Karl Fischer titration. Compared to other isotherms (e.g. as depicted in the results above seem to have much too steep gradient at low moisture contents (below RH = 5 %). This does not necessarily mean an error. On the contrary, the data in [Du 1999] are only extrapolated in the important relative humidity range below 20 %, [Oommen 2008]. Additionally it must be emphasized, that each wood based material has a unique isotherm. [Tveit 1966] found very different results for various cellulose materials. Another explanation for the steep gradient might be that no discrimination between adsorption and desorption was made. Desorption curves are steeper close to the origin than adsorption curves (Figure 19).

Since the capacitive probes used here are carefully calibrated with salt solutions and compared to chilled mirror dewpoint hygrometers (p. 31), their tolerance comprises ± 0.2 % relative humidity in the humidity range of 2-80 %. Thus they

feature a high accuracy at least at room temperature. At elevated temperatures the accuracy limits are below ± 2 % relative humidity [Vaisala 2002].

For the Karl Fischer instrument the situation is worse. Any accuracy can hardly be given because of the lack of instruments having higher accuracy. Although periodic checks with standard water samples conditioned the instrument, the round robin tests, as depicted in Figure 8, attest a poor comparability between different laboratories. Because in this investigation a heating temperature of 160-180°C and heptane as oil solvent were used, the results were somewhat higher compared to the moisture by weight results from other laboratories. Figure 10 may help to show the deviation to be distinctly below 0,7 % moisture by weight.

3.3 Solubility of Water in Oil

3.3.1 Composition and Aging of Insulation Oil

Mineral Oil

A mineral transformer oil consists mainly of hydrocarbons of three structures: paraffin, naphthenes and aromatic. Paraffinic structures are straight or branched molecules and show a lower thermal stability than the other structures. The paraffinic type of molecule is nonpolar and has a low affinity for water and polar aging products. Naphthenic molecules form ring structures and have better low temperature properties. The aromatic molecules are ring-structured too with double and single bonds. They greatly influence the oil properties. For example they have a slight polarity and are therefore responsible for the water solubility of the corresponding oil (Table 3). Furthermore they inhibit the oil against oxidation [Nynas 2004].

Mineral oils age because of oxidation. The two main parameters influencing oxidation are oxygen and temperature, the metals acting as catalysts. The oxidation process occurs in several steps and produces by-products: radicals, alcohols, water, aldehydes, ketones, carboxylic acids and finally esters, [Nynas 2004]. Some of the by-products are polar giving losses in the electrical field. Some substantially increase the water solubility of aged oils.

Composition and Aging of Vegetable Oil

Vegetable oils are derived from plants and composed of esters. The esters are glycerol and one or more saturated or unsaturated fatty acids. The degree of saturation determines the oxidation stability of the oil. On one hand a saturated fatty acid is stable but has a high freezing point, on the other hand the di- or triunsaturated acid is more susceptible to oxidation. A mono-unsaturated acid is the best choice according to [Oommen 1997]. Oxidation inhibitors make the oil more stable. Vegetable oils also deteriorate because of hydrolysis, thus, in contrast to mineral oil, a high water saturation harms the oil.

Water in Oil

The water solubility of mineral oil depends on the temperature and the amount of aromatic hydrocarbons and polar molecules in the oil. These amounts are affected by the refining process and oil aging. Water exists in the following states:

- Dispersed as water vapour The polar water vapour molecules are dispersed between the nonpolar oil molecules as a gas [Gerrard 1980].
- Attracted in oil by aromatic components or aging by-products, Every component able to coalesce hydrogen or Van der Waals bonds with water molecules will increase the water solubility of the oil.
- Adsorbed by cellulose fibres
- As free water If the oil is supersaturated, the water molecules will form droplets or clusters with the typical hydrogen bonds between molecules.

Vegetable oils in contrast to mineral oils contain fatty acids which enable them to dissolve 20-50 times more water than the saturated hydrocarbons of mineral oils. In vegetable oils the water molecules are attracted to the OH-groups of the fatty acid molecules.

3.3.2 Measurement of Isotherms at Insulation Oils

Investigated Oils and Measurement Setup

The research focused on mineral oils made by Shell and Nynas since these companies manufacture the most commonly used insulation oils. New mineral oils explored were Shell Diala D, Nynas Nytro 3000 and Nynas Nytro 3000X (inhibited). Aged oils were a 25 years service-aged with a neutralization number of 0,13 mg KOH/g oil and a service-aged Shell K 6 SX from 1965 with a neutralization number of 0,49 mg KOH/g oil. Additionally four vegetable oils were investigated; Envirotemp FR3, Midel eN, sunflower oil and the synthetic ester Midel 7131.

The temperature range comprised 20-80°C, which is usual for power transformers. To increase the accuracy the graphs are superpositions of several measurements. Two capacitive probes (Vaisala HMP 228) as described in section 2.3 measured the relative saturation directly in the oil. To ensure a fast equilibration between the liquid and the capacitive probe the vessels were mechanically shaken (Figure 23). Coulometric Karl Fischer titration with direct injection into the reaction vessel (ECH Aqua 40.00) determined the moisture by weight in ppm following the procedure of section 7.1. The relation between

moisture relative to weight and moisture relative to saturation is given by moisture isotherms. Thus the saturation water content at a specific temperature was calculated from the reading of the capacitive probe (relative saturation) and the corresponding titration result (water relative to weight).



Figure 23: A two-litres oil vessel with a capacitive probe inserted in the shaking mechanism

Isotherms in Insulation Oil

The isotherms in oil relate moisture relative to weight W (ppm) to the relative saturation RS of the oil at a specific temperature. Two differences arise compared to isotherms in cellulose:

- The absolute water content in oil is linearly proportional to the water vapour pressure up to the saturation level. This is typical for ideal gases dissolved in ideal liquids, were no chemical reactions happen, [Gerrard 1980]. However, with increasing amount of impurities a nonlinear behaviour according to the theory of [Brunauer 1940] may appear similar to Figure 16.
- At increasing temperature the moisture solubility in oil *increases* while that of cellulose *decreases*. At higher thermal energy more water molecules are able to enter the gaseous state and to remain in it, therefore more molecules are dispersed as water vapour in the oil.



Figure 24: Moisture sorption isotherms for various oils: synthetic ester Midel 7131, natural ester Envirotemp FR3, new Nynas Nytro 3000 and service-aged 100 Shell Diala K 6 SX at 20 and 80°C

Figure 24 illustrates the moisture sorption behaviour of insulating liquids. Aging clearly increases the moisture solubility of mineral oil. The synthetic ester Midel 7131 shows an exponential increasing water solubility, indicating a non-ideal water adsorption caused by at least two different moisture uptake mechanisms. All other liquids behave like ideal liquids, where the water content relative to weight is linearly related to the partial pressure of its water vapour.

Since moisture relative to weight and moisture relative to saturation are simply linearly related to each other in an ideal liquid, it is more usual to use moisture saturation diagrams. They effectively depict the moisture solubility of oils as a function of temperature (Figure 25 and Figure 26).



Figure 25 depicts the saturation graphs for mineral oils. Aging obviously increases the moisture solubility because of aging by-products as described above. However, the lower molecular acids like formic and acetic acids rather than the overall acidity (total acid number in mg KOH per g oil) increase the water solubility in oil. Lower molecular acids originate from paper decomposition [Lundgaard 2004], whereas the higher molecular acids (e.g. stearic acid) come from oil oxidation.

Figure 26 shows the much higher moisture solubility in vegetable oils due to water attraction by polar groups of the esters. All natural esters tested show a very similar water solubility. They consist of three ester groups, whereas the synthetic ester Midel 7131 has four ester groups, which result in a higher solubility.



Water Solubility Coefficients

The temperature dependence of the saturation solubility of water in oil W_s can be expressed by an empirical equation:

$$W_{\rm s} = A \cdot e^{-B/T} \tag{4}$$

A and B are constants, which are similar for new mineral transformer oils but differ with aromatic contents and water attracting components. Table 3 below lists the coefficients for equation (4) and also illustrates the influence of aromatic compounds. Since the three natural vegetable oils show similar behaviour, the coefficients for only one of them is given (Envirotemp FR3, Midel eN and sunflower oil).

			Water solubility in ppm			
Oil type	А	В	20°C	40°C	60°C	80°C
5 % Aromatics ¹	$16,97 \cdot 10^{6}$	3777	42,8	97,5	201	382
21 % Aromatics ¹	$13,16 \cdot 10^{6}$	3538	75	162	319	584
Shell Diala D ¹	$24,65 \cdot 10^6$	3835	51	117	245	471
Nynas Nytro 3000	$8,76 \cdot 10^{6}$	3504	56	120	236	428
Aged Diala K 6 SX	$1,84 \cdot 10^{6}$	2752	153	264	428	725
Envirotemp FR3	391278	1803	832	1232	1742	2367
Midel 7131	349505	1457	2420	3325	4398	5635

 Table 3:
 Water Solubility Coefficients and solubility for various insulation oils

¹ [Sokolov 2008]

Accuracy of the Results

The measurement of the relative moisture saturation and moisture relative to weight determine the accuracy of the moisture saturation graphs. Figure 25 especially reveals a distinctly higher saturation level compared to the literature.

The capacitive probes used here provide sufficiently high accuracy, but are limited by diffusion effects at low temperatures and low water concentrations. If a wet probe with many water molecules attracted is inserted into a dry oil, it takes a very long time before the probes water molecules become dissolved in the oil and equilibrium is established. This may also result in overly wet oil locally surrounding the capacitive probe. To diminish this error a mechanism shook the oil vessel. Hence the error is considered to be less than -4 % relative saturation at low temperatures and this for mineral oils only. The usual error of the capacitive probe of $\pm 2\%$ relative saturation results in a deviation of -12 to +18% of the ppm saturation result, i.e. the saturation results in Figure 25 and Figure 26 have this error range. In the worst case, Midel 7131 at 80°C having a relative saturation of 3,5%, the ppm saturation result has the broad error range of -29 to +160%. On the other hand the results for Midel 7131 correspond very well with data from the literature.

The Karl Fischer titration decreases the accuracy even more. The round robin test in section 2.1 has disclosed the very poor comparability between different laboratories especially for oils. A percent deviation cannot be given. Fortunately the improved moisture measurement approach introduced in section 3.5 eliminates the need for Karl Fischer titration of oil and for moisture in oil isotherms.

3.4 Moisture Diffusion and Equilibrium

Thermodynamic and Moisture Equilibrium

Thermodynamic systems tend to reach equilibrium because this is the state with the highest entropy. A system is in thermodynamic equilibrium, if its properties do not change and all have fixed values [Cengel 2006]. The types of equilibrium depend on the conditions of the system, for power transformers they are:

- Thermal equilibrium No temperature differentials exist throughout the whole system, thus no heat flow occurs.
- Mechanical equilibrium A system is in mechanical equilibrium if pressure does not change with time but may change with place due to gravitational effects.
- Phase equilibrium The mass of each phase reaches an equilibrium level and stays there.

• Chemical equilibrium The chemical composition does not change with time, no chemical reactions occur.

All four general equilibrium criteria influence moisture equilibrium and cause diffusion as the result of non-equilibrium. Temperature differences have the main impact on moisture diffusion, since water solubility in oil and water adsorption in cellulose change with temperature. With increasing temperature the potential to absorb water in cellulose decreases while that of oil increases. Thus the equilibration forces water molecules to migrate from cellulose to oil, whilst for decreasing temperatures it is the other way round.

Pressure as the driving force for mechanical equilibrium causes for example the typical breathing of open conservator systems, increasing the moisture content in the transformer.

Phase equilibrium applies for example to water vapour and water droplets, but is of minor importance.

Chemical equilibrium plays a role in water accelerated aging processes such as in the hydrolysis of cellulose.

In a power transformer a global thermodynamic equilibrium will hardly ever be reached because of the varying load factor and long time constants. During service homogenous temperature and pressure distribution is impossible. Nevertheless a local thermodynamic equilibrium will occur. Here the macroscopic observables are varying in space and time, but so slowly, that for any point one can assume thermodynamic equilibrium in some neighbourhood about that point. This local equilibrium will occur if a transformer is operated at a constant load for a sufficient time. Therefore moisture in oil and paper are in local equilibrium. At low temperatures even a local equilibrium won't exist (p. 51).

Water Potential, Vapour Pressure and Relative Humidity

Moisture equilibrium in a two phase system is set, if water potential of the gaseous phase Ψ_{gas} is identical to that of the adsorbed phase Ψ_{ads} [Myer 1965].

$$\Psi_{gas} = \Psi_{ads} \tag{5}$$

Water potential describes the thermodynamic state of water in materials and can be given in units of pressure or energy. Moisture diffusion tries to establish the same thermodynamic potential of water in all materials of one system. Water potential depends on temperature, solute potential (term with water vapour pressure in equation (6)), pressure and matrix effects such as cohesion and surface tension. Equation (6) defines the thermodynamic potential in the gaseous phase. Ψ_0 here is a standard potential, R the ideal gas constant [Murrel 1982]. For moisture diffusion between parts of the insulation system the most important variables are temperature and water vapour pressure.

$$\Psi_{gas} = \Psi_0 + R \cdot T \cdot \ln\left(\frac{p}{p_s}\right) \tag{6}$$

Since the equation above includes the definition of relative humidity as p/p_s (p. 136), moisture exchange can also be described in terms of relative humidity or relative saturation: Equilibrium processes result in identical moisture relative to saturation in adjacent materials, supposing the same temperature and pressure apply (7). The material might be cellulose, oil, air or even a plastic.

$$RS_{Cellulose} = RS_{Oil} = RH_{Air}$$
⁽⁷⁾

Relative saturation is equivalent to water activity, so moisture exchange can be expressed also in terms of water activity (8), [Roizman 2005].

$$a_{W,Cellulose} = a_{W,Oil} = a_{W,Air}$$
(8)

Since p_s in equation (6) is the saturation pressure in a gas, moisture migration can also be expressed in terms of equal water vapour pressure p. Hence differences in water vapour pressure force moisture migration, (9).

$$p_{Cellulose} = p_{Oil} = p_{Air} \tag{9}$$

The different quantities reflect the different descriptions used in the literature.

Practical Verification of the Equilibrium Theory

A practical test not only confirmed the theory about moisture equilibrium in transformer materials, but also proved the applicability of the commercial instruments. This is even more important, since the theory lays the foundation for the later improved measurements by moisture equilibrium.

67 g of non-impregnated new Kraft paper having 2,85 % moisture by weight filled a two-litre-vessel (Figure 23). The relative humidity of the air in the vessel was measured with capacitive probes during 3-5 temperature cycles of 25-40-60-80-25°C. Then the air was exchanged for oil and the relative saturation in oil measured during the same temperature cycles. In a last step the oil was removed and the relative humidity for the now oil-impregnated Kraft paper measured. According to the theory above, under all conditions the relative humidity of the air and the relative saturation of the oil should remain identical.

Figure 27 illustrates one heating cycle, showing temperature and relative humidity over non-impregnated Kraft paper. The equilibration process occurs rather fast because of the large surface of the non-impregnated Kraft paper. At 80°C the time to reach equilibrium is 1,9 h, at 27°C it is 3,6 h. For the relative



saturation measurement in oil the time to reach equilibrium at 27°C was somewhat longer at 5,5 h.

Figure 28 shows the relative humidity or relative saturation as a function of temperature for all the three conditions, non-impregnated, oil-immersed and impregnated Kraft paper. The result show that during all these conditions the relative humidity remained constant at 8-18 % RH. Results of 30 and 35 % RH are the starting values at non-equilibrium. The slight increase of the RH and RS-readings from 8 to 15 % at 26°C was caused by a leak in the test vessel; proved by the fact that moisture by weight increased from 2,85 to 3,35 %.

This test confirms the statement above, that equilibrium processes result into an identical moisture relative to saturation in adjacent materials, supposing the same temperature and pressure rules. It also shows that a reproducible

equilibrium cannot be achieved at room temperature but only several heating cycles leading to a repeatable reading.

3.5 Improved Measurements by Moisture Equilibrium

Section 3.1 has shown that the conventional application of equilibrium diagrams usually results in erroneous diagnosis results. The materials moisture adsorption capacity and aging make the use of curves based on ppm from the literature impossible (Figure 15). The next sections introduce new approaches based on moisture equilibrium in order to reliably evaluate moisture content in the solid insulation.

3.5.1 Diagrams Adapted to the Moisture Adsorption Capacity

The first step to improve equilibrium diagrams is to adapt them to the water adsorption capacity of the involved materials.

Creating Equilibrium Diagrams

To measure moisture in cellulose and oil and make equilibrium diagrams one can use direct or indirect moisture measurement.

In the direct method oil and cellulose are in contact with each other and the cellulose is oil immersed. The measurements take part in oil and cellulose samples at various temperatures after equilibrium is reached. Karl Fischer titration determines the moisture content. Drawbacks of this method are the long time constants to establish equilibrium especially at lower temperatures and the unreliability of the Karl Fischer method (section 2.2).

The indirect method is based on the physical law which states that the relative saturation for adjacent materials is equal under equilibrium conditions (see above). The ambient medium could be air or oil, supposing they are at the same temperature and pressure. Then moisture isotherms for cellulose materials and for oils are combined by equating the x-axis (relative saturation). This method was firstly applied by [Oommen 1983].

Advantages of this method are, that equilibrium in oil free cellulose is established much faster compared to oil immersed cellulose, the moisture profile becomes more homogenous, the measurement of moisture relative to saturation is much easier and continuously possible compared to moisture relative to weight in ppm by Karl Fischer titration.



Figure 29: Illustration for the indirect method to create an equilibrium diagram by equating the x-axis of moisture isotherms for oil and cellulose

Examples of Equilibrium Diagrams

Using the described approach many combinations of isotherms are possible. Here only a few practical cases are depicted: new Nynas Nytro 3000 with new Kraft paper and new pressboard, service-aged Shell K6SX with thermally degraded Kraft paper and service-aged pressboard and, finally, new synthetic ester Midel 7131 and new vegetable oil Envirotemp FR3 with new Kraft paper.



Figure 30: Equilibrium diagram for new mineral oil Nynas Nytro 3000X with new Kraft paper and new pressboard



Figure 31: Equilibrium diagram for service-aged mineral oil Shell K6 SX with thermally degraded Kraft paper and service-aged pressboard

Figure 30 depicts equilibrium diagrams for new Kraft paper and new Nynas Nytro 3000X and for new pressboard and new Nynas Nytro 3000X. Under new conditions the difference in the moisture saturation level for Nynas Nitro 3000, Nynas Nitro 3000X and Shell Diala D is small, thus one set of curves is valid for all oils. Ageing shifts the curves to the right, since the moisture solubility in oil increases (Figure 31).



Figure 32: Equilibrium diagram for new vegetable oil Envirotemp FR3 and new synthetic ester Midel 7131 with new Kraft paper

Figure 32 illustrates the much higher moisture solubility in vegetable oils and the equilibrium curves move toward oil.

Water Distribution Between Oil and Cellulose

Based on the equilibrium diagrams above one can calculate the moisture distribution between cellulose and oil under equilibrium conditions. Here a new, dry transformer, an aged, wet transformer and an oil change in that wet transformer by a natural vegetable oil were considered. New conditions here mean that Nynas Nytro 3000 together with new cellulose material has 0,6 % water relative to weight (Figure 30). Aged and wet mean an aged Shell K6SX with service-aged pressboard having 3 % water relative to weight (Figure 31). Finally the oil exchange at the aged transformer should be done with Envirotemp FR3. This example of a transformer has 7 t of cellulose and 70 t of mineral oil, which is a usual ratio for large power transformers. Because Envirotemp FR3 has a higher density, the fluid mass will be 75 t after exchange. As average insulation temperature 40°C is assumed. Table 4 and Figure 33 depict the water distribution between oil and cellulose in the three mentioned cases.

	New, dry		Aged, wet		Aged, oil exchange	
	Cellulose	Nytro 3000	Cellulose	K6SX	Cellulose	New FR3
Insulation mass	7 t	70 t	7 t	70 t	7 t	75 t
Water content	0,6 %	2 ppm	3 %	44 ppm	2,84 %	190 ppm
Water mass / kg	42	0,14	210	3,1	199	14,2

Table 4:Water distribution between oil and cellulose under new and aged
conditions and after oil exchange



Figure 33: Water distribution between oil and cellulose under new conditions, aged and wet conditions and after oil exchange by a natural ester

Figure 33 conclusively shows that cellulose contains the major part of water in a power transformer. Cellulose serves as a water storage medium, whereas oil serves for water transfer. The moisture distribution ratio is substantially changed only by introducing an insulating liquid with a high water solubility, as here the natural ester Envirotemp FR3.

Hence transformer drying can't be done by oil exchange only, rather by oil circulation in a drying system for a long time. For this purpose an oil with a high water solubility serves as a better drying medium.

Need for Further Improvements

Using the equilibrium diagrams depicted above it is possible to determine the "true" water content in cellulose, since they are adapted to the moisture absorption capacity of the materials. They still have the essential drawback, that their validity is restricted to the involved materials. For other materials and aging conditions in the particular transformer on-site they have to be adapted. Thus the next step to *more universal* equilibrium diagrams is described in the following subsection.

3.5.2 Measurement via Moisture Saturation of Oil

In this approach instead of moisture in oil relative to weight (ppm) the relative saturation in oil (%) is used. Additionally the diagrams are adapted to the moisture adsorption capacity of the cellulose. The method is based on the thermodynamic law, that moisture relative to saturation is the same for materials in contact with each other under equilibrium conditions (equation (7)). Thus it is possible to simply use the moisture adsorption isotherms as depicted in Figure 20, Figure 21 and Figure 22 to derive moisture relative to weight in cellulose from moisture relative to saturation of the surrounding medium. In this application the surrounding medium is not air but oil, but the same thermodynamic law is valid. This approach results into the following advantages:

- Oil aging and its influence on moisture saturation level becomes negligible as it is already included in the measurement of moisture saturation.
- With relative moisture on the X-axis the graphs become less temperature dependent compared to moisture by weight on that axis.
- On-site and on-line application is possible.
- Load cycles can be equilibrated with a long term average.
- Errors due to sampling, transportation to the lab and titration are excluded. The uncertainties of capacitive probes are insignificant compared to the excluded influence.
- Capacitive sensors measure the relative moisture in oil continuously and are easy to integrate into a monitoring system.

Figure 34 illustrates the moisture determination using adsorption isotherms for new and thermally degraded Kraft paper. With thermal aging the ability of

cellulose to adsorb moisture decreases. With the reading of a capacitive probe (here 10 % relative to saturation and 60°C oil temperature) the moisture content in Kraft paper can easily be determined (here under new conditions 3 % relative to weight, for thermally degraded paper 2,8 %). To balance load cycles, an average of extended time measurements will provide more reliable results.



Figure 34: Determination of water content in new and thermally degraded Kraft paper via moisture sorption isotherms

The approach to using moisture saturation of oil substantially improves moisture determination in transformers. Still the diagrams have to be adapted to the moisture adsorption capacity of the specific cellulose material. Moreover, if someone estimates the moisture in a transformer, the use of moisture relative to saturation for cellulose is also more meaningful than the traditional way which is to use moisture relative to weight. The next paragraphs will explain this approach in more detail.

3.5.3 Measurement of Moisture Saturation in Cellulose

Correlation of Moisture Saturation to Destructive Effects

Moisture saturation is a critical factor that determines the amount of water *available for interaction* with materials. The destructive effects of water in power transformers decreases the dielectric strength of oil, accelerates aging of cellulose and causes bubble formation at elevated temperatures.

All these destructive effects cause water molecules, which are available for interactions with materials, to be active. These water molecules enable acids to dissociate and thus to provide charge carriers for breakdown processes. This clearly illustrates Figure 2 on page 14.

These molecules are also responsible for aging of cellulose by hydrolysis. The dependence of this aging process on relative saturation is illustrated in Figure 4. Here the expected life for the solid insulation does not linearly depend on the

water content relative to weight. The graph indicates the relation of water content relative to weight and saturation via the s-shaped moisture sorption isotherms of cellulose materials (Figure 19). Most reaction rates increase with increasing water saturation and correlate better with water saturation than with moisture content [Bell 2002]. Moisture sorption isotherms are useful to relate water to the moisture-induced chemical and physical deterioration of materials.

Furthermore the evolution of gas bubbles because of evaporation depends on the evaporable water, not on strongly bound water [Koch 2007.2].

Water relative to weight, measured by e.g. Karl Fischer titration, unfortunately reflects the bound and less active water as well. Water molecules that are strongly bound in a monolayer are not available for further interactions. Thus moisture relative to saturation - not relative to weight - determines the available water for destructive effects.

Using the relative moisture saturation of oil *and of cellulose* gives the following advantages:

- Continuous, accurate measurement with capacitive sensors,
- Easy implementation in monitoring systems,
- Neither oil nor paper aging effects the accuracy of the measurement,
- Conversion via equilibrium charts is no longer necessary,
- Water saturation indicates water molecules removable by drying,
- Direct relation to the destructive effects of water.

Practical Implementation

Figure 35 illustrates the application of a relative saturation measurement using a capacitive probe in a power transformer equipped with an on-line monitoring system. The load factor influences the top oil temperature, which follows in diffusion processes changing the relative saturation in oil. A long term average equilibrates the relative saturation in oil with the relative saturation of the surrounding cellulose and comes to 4,1 %. Obviously this result applies for the cellulose and oil that are in equilibrium. Through the long time average a mathematical equilibrium is reached, balancing the daily load cycles of a transformer. With these few steps to obtaining moisture relative to saturation in oil *and cellulose* a reliable and meaningful moisture determination in power transformers is completed.



Additionally, by a moisture isotherm as Figure 34 one can derive moisture by weight in cellulose as well which is 2,2 % in this case. [Tenbohlen 2004] applied this method in a preliminary manner.

Consideration of Temperature Zones

The results obtained gain in reliability if the probe is inserted into the hot oil flow before the cooling system or as close as possible to the winding. Figure 36 clearly depicts the temperature distribution in a large power transformer with natural cooling according to [IEC 60354] based on an ambient temperature of 20° C.



The goal is to measure relative saturation and temperature of the insulation paper in the windings, since these insulation parts determine the moisture content of the oil, are most subject to aging and may emit bubbles. The insulation paper will have a temperature between that of the surrounding oil (5472°C) and the conductors of the winding (73-91°C). A capacitive probe installed in the hot oil flow on top of the transformer will measure the top oil temperature (72°C) and its corresponding relative saturation. Thus it will give a good average of the relative moisture saturation of the insulation paper.

The pressboard insulation structures in the main duct between high and low voltage winding will not be considered. Because of their lower temperature around average oil temperature (64°C) they will conduct water. These rather wet structures will not be reflected in the relative saturation measurement. On the other hand, relative saturation of cellulose does not greatly depend on temperature (Figure 20). The result, valid for winding paper, might be increased somewhat to include the colder structures as well. Finally these insulation structures are not as relevant to the destructive effects of moisture as the paper in the winding.

Accuracy of the Relative Saturation Measurement

The accuracy of the approach to measure relative saturation in oil and cellulose depends on the capacitive probe, the installation conditions and the equilibrium process. An uncalibrated probe already provides an accuracy of ± 2 % in the whole temperature range, which is sufficient for moisture assessment. A periodical check of the calibration, e.g. every two years, will insure a proper result over long time periods.

The location of the probe causes a greater impact on the accuracy. An installation at the bottom of the cooling system will give a too low temperature and – even worse – too high moisture saturation. An installation in the hot oil flow coming from the winding ensures high reliability of the result.

One single on-site measurement in an oil sample without continual measurement storing and averaging in a data logger suffers from the current state of equilibration. Under non-equilibrium of moisture in oil to moisture in cellulose the result will be wrong. Fortunately the on-site measurements at power transformers show good agreement with other results even under these single sampling conditions (p. 107).

Chapter 3 introduces moisture in oil and paper and describes the measurement technique via moisture equilibrium. The following chapter 4 elaborates another indirect approach to measuring moisture in transformers, based on the dielectric conditions of the insulation materials.

4 Moisture Analysis by Dielectric Methods

"Dielectric response measurements provide valuable information on the state of oil-paper insulation in power transformers, in particular on moisture content." [Gubanski 2004]

Dielectric diagnostic methods deduce moisture in paper or pressboard from dielectric properties like return voltage, polarisation and depolarisation currents and dissipation factor. The investigations presented here are intended to improve the reliability of moisture determination.

This chapter is structured into six sections: It starts with a literature review and a motivation for the present research work. The second section deals with the theoretical background of the dielectric properties of materials. Thirdly the dielectric properties of oil are measured, fourthly that of cellulose in time domain, and fifthly in frequency domain. Finally the sixth section describes an improved moisture analysis algorithm.

4.1 State of the Art

4.1.1 Literature Review

Primary motivations for the development of dielectric response methods were the lack of methods for on-site moisture assessment in power transformers and the disappointing results of the hitherto used conventional equilibrium approach (section 3.1). The methods developed later gave promise of higher accuracy and were designed for on-site application.

Recovery Voltage Method RVM

Measurements in time and frequency domain can display the properties of a dielectric. Time domain voltage measurements are applied as recovery voltage measurements after charging the insulation with a DC voltage. After charging time t_C a short circuit will be applied for t_d and then the peak recovery voltage $U_{r,max}$ and the time to peak t_{peak} recorded (Figure 37). By subsequent relaxation and repeated charging for varied times a so called "polarisation spectrum" can be created (Figure 41).



Figure 37: Procedure of a recovery voltage measurement

At first, [Nemeth 1991] introduced the recovery voltage method. Its interpretation method derives moisture in cellulose from the polarisation spectrum. Soon users questioned the reliability of the moisture result [Kachler 1996]. Subsequently two additional dielectric response methods were developed: the polarisation and depolarisation currents PDC and the frequency domain spectroscopy FDS, [Houhanessian 1998] and [Neimanis 2000].

Polarisation and Depolarisation Currents PDC

A time domain current measurement records the charging and discharging currents of the insulation. They are usually called Polarisation and Depolarisation Currents PDC. Figure 38 illustrates the measurement of the dielectric properties of an insulation by polarisation and depolarisation currents using linearly scaled axes.



Figure 38: Polarisation and depolarisation currents during charging by a DC voltage U_C for a time t_C and after short-circuiting

Frequency Domain Spectroscopy

Frequency domain measurements are derived from old known dissipation factor measurements, yet with a frequency range especially enhanced for low frequencies. The derived measurement method is called Frequency Domain Spectroscopy FDS. Figure 39 shows the s-shaped curve of the dissipation factor via frequency as typical for a power transformer.



Figure 39: Dissipation factor while applying a sinusoidal voltage of varying frequency

The CIGRÉ task force 15.01.09 compared the three methods and concluded: "For the RVM technique, the old interpretation based only on simple relationship between the dominant time constant of the polarisation spectrum and the water content in cellulose is not correct" [Gubanski 2004]. In contrast to this [Saha 2003] attested it to be "sensitive to aging and moisture". This was the situation at the beginning of the research work presented here. Furthermore it was, of course, of interest to see how the newer methods PDC and FDS performed. Thus a systematic investigation of the three competing methods was carried out in order to reveal their strengths and weaknesses and to get a starting point for subsequent improvements. There are two additional reasons for research.

Influence of Conductive Aging Products

Some researchers found, that aging increases the dielectric losses and thus has a similar influence as water. [Ekanayake 2006] and [Shayegani 2005] have already investigated this effect, but gave no approach to compensate for it. This topic is also alluded to in a paper describing the work of Cigré on power transformers [Guuinic 2001]. In data analysis these aged transformers appear more moist than they really are. This also illustrates a field measurement at p. 114. This thesis investigates this influence and also shows a way to compensate for it.

Time Dependent Oil Conductivity

Frequently discussed topics are the time and voltage dependent characteristics of insulation oil. [Kuechler 2004] and [Koch 2005] reported, that particularly aged oils show decreasing oil conductivity with increasing test time. This behavior complicates the application of a linear model for the dielectric properties of materials. Section 4.3 discusses this issue.

4.1.2 A Comparative Test on the State of the Art Methods

The commercial dielectric response methods for moisture determination at power transformers were compared in order to give a basis for further improvements. Three instruments with corresponding analysis software were available: the Recovery Voltage Meter RVM 5462 by Haefely Tettex with analysis software SWRVM 2 V.3.0, [Haefely 2007], the Polarisation Depolarisation Currents Analyser MOD1 by Alff Engineering with analysis software PDC Evaluation Software V.3.0, [Alff 2007] and the Insulation Diagnostics System IDA 200 from GE Energy Services with analysis software MODS V.1.5, [GE 2007]. It should be pointed out that data analysis depends on the applied software, which was in this comparison delivered together with the instruments.

Description of the Test Model for the Systematic Comparison

A large insulation model called "Pancake Model" served to benchmark the three methods. A variation of the following parameters was investigated: insulation geometry, insulation temperature and oil conductivity. In the ideal case the analysis methods should be able to compensate for these three parameters. During all investigations the moisture content in cellulose remained constant at 1,0 %, measured for paper and pressboard samples with coulometric Karl Fischer titration at 160°C heating temperature.





The model consists of eight pancake shaped coils with oil ducts between them (Figure 40). The ratio of barriers and spacers to oil ranged from 15 to 100 % as described in the following table. This simulates the main insulation of different transformers. The dielectric properties were measured at 21°C, 50°C and 78°C insulation temperature and once again at 21°C. During the temperature investigations a new insulation oil type Shell Diala D (conductivity 1,6 pS/m) filled the model tank. This oil was exchanged by a 25 years service aged transformer oil (conductivity 16,5 pS/m) and the measurements were repeated at 21°C.

Connection	Oil / Barriers	Oil / Spacers	Illustration of oil/pressboard ratio
CH – B	83 / 17	85 / 15	
DG – CH	72 / 28	72 / 28	
E – DG	50 / 50	45 / 55	
F-E	0 / 100	0 / 100	

 Table 5:
 Ratio of oil to cellulose in the insulation model

Although the pancake model has conditions similar to power transformers, there is still a remarkable difference. The insulation geometry is constructed in the shape of disks, but at core-type power transformers in the shape of cylinders. This might increase the influence of parallel currents on the measurement results. Beside this the geometrical condition F-E (100 % pressboard) contains small oil ducts close to the paper-wrapped conductors.

Analysis Results of RVM, PDC and FDS

Figure 41, Figure 42 and Figure 43 display on the left hand side the common interpretation scheme of the specific method. The graph on the right displays the moisture content as analyzed by the software belonging to the three commercial instruments. The dotted line marks the moisture content as measured by Karl Fischer titration.



Figure 41: The RVM interpreted by the "polarisation spectrum" (left) and moisture analysis results with SWRVM 2 V.3.0 (right)

The results of the RV analysis software in Figure 41 strongly depend on oil conductivity. After oil exchange the highest moisture content was analyzed, although the moisture content of paper was unchanged during the

measurements. Temperature and insulation geometry influence the results as well. Thus the software SWRVM 2 V.3.0 using the "polarization spectrum" can't clearly determine moisture in oil-paper-insulations, it does not compensate for oil conductivity and only poorly compensates for insulation temperature.



Figure 42: Interpretation scheme for the PDC method (left) and analysis results with PDC Evaluation Software V.3.0 (right)

PDC analysis results show a small sensitivity to insulation geometry and a slight temperature dependence, as shown in Figure 42. With increasing oil conductivity the evaluated moisture content increased, although in reality it remained constant. Nevertheless the analysis results are very close to the moisture content measured by Karl Fischer titration.



Figure 43: Interpretation for the FDS method using dissipation factor (left) and analysis results obtained with software MODS V.1.5 (right)

FDS analysis provides the best compensation of insulation geometry. However with increasing temperature the cellulose seems to dry, which actually happens because of moisture diffusion, but not in this dimension. This tendency actually reveals an imperfect temperature compensation. Similar to the other methods the increased oil conductivity increases the moisture analysis results as well, although moisture content in the cellulose remained constant.

To sum up, this comparative test found:

- The commercial evaluation software for PDC and FDS are able to compensate for the influences of insulation geometry, insulation temperature and oil conductivity.
- The commercial analysis software for the RV method based on the central time constant is too simplistic.
- All methods show an apparently *decreasing* moisture analysis result for increasing temperature and *increasing* moisture result for increasing oil conductivity.

This work tries to improve these weaknesses of the benchmarked methods by introducing a more comprehensive analysis algorithm on p. 105.

4.2 Polarisation and Conductivity of Insulation Materials

The multilayer oil-paper insulation of power transformers consisting of oil and paper shows polarization and conductivity phenomena. Dielectric diagnostic methods measure the interfacial polarization effect that originates from the interfaces between cellulose and oil. Polarization is superimposed by the DC conductivity of cellulose and oil. Moisture, temperature and conductive aging products change these phenomena.

Section 4.2 gives an introduction to the theoretical explanation, to modelling and measurement of dielectric properties. It is based primarily on the work of other researchers.

4.2.1 Dielectric Properties – Theoretical Background

Electric Displacement and Polarisation of Materials

Polarisation appears, if a material is exposed to an electrical field, e.g. [Jonscher 1996]. At first let us consider a capacitor having a homogenous field with vacuum as an insulator. Here the electric displacement field D is proportional to the electric field vector E with ε_0 as relating constant.

$$D = \varepsilon_0 E \tag{10}$$

Now a plate of dielectric material will replace the vacuum. Inside this plate the electric field will cause a polarisation P on an atomic scale. The electromagnetic susceptibility χ relates polarisation to the electric field.

$$P = \chi \varepsilon_0 E \tag{11}$$

The susceptibility χ is zero for pure vacuum but increases because of polarisation processes in a solid dielectric material. The relation to dielectric permittivity ε is given by

$$\varepsilon = \varepsilon_0 \left(1 + \chi \right) \tag{12}$$

The polarisation P results in opposite charges at the surface of the dielectric followed by an opposite field. This field contributes to the electrical displacement.

$$D = \varepsilon_0 E + P = \varepsilon_0 (1 + \chi) E \tag{13}$$

(13) indicates that a polarisation can also exist without an external electrical field. This is the case after charging of a dielectric, where the depolarisation process causes an electric displacement. The different polarisation processes have different time constants, resulting in a time- and frequency dependent susceptibility χ .

Polarisation Mechanisms and Frequency Dependence

Depending on the molecular and geometric conditions of the particular dielectric various polarisation mechanisms can appear [Münch 1987]:

• Electronic polarisation

Here the electric field distorts the orbit of electrons surrounding the atomic cores. This effect applies to all materials and because of the small dimensions it is effective up to optical frequencies.

• Ionic polarisation

The electric field induces net dipoles by slightly displacing the ions from their rest position. This is usual for salts and works up to frequencies of the infrared light.

• Dipolar polarisation

Molecules with permanent dipole moments follow the field up to some mega or giga Hertz.

• Interfacial polarisation

This effect is typical for inhomogenous dielectrics with different permittivity or conductivity. Here charge carriers such as ions become deposited at the interfaces forming clouds with a dipole-like behaviour. This kind of polarisation is effective only below some ten Hertz. It is the most important effect in the context of dielectric diagnostic methods. • Hopping or trapping of charge carriers

Here charge carriers as electrons and ions are locally trapped by charged sites. Charge transport cannot progress smoothly and appears on the outside as a polarisation mechanism. This appears e.g. in Polymers especially at high temperatures and very low frequencies [Jonscher 1996].

The interfacial polarisation is the most important polarisation process in the context of this work. Moisture in paper and pressboard influences this process in superposition with conductivity phenomena.

4.2.2 Dielectric Properties in Time Domain

Current Density and the Dielectric Response Function

To cover the physical phenomena of relaxation and conductivity currents with mathematical expressions, one may start with the famous equations of Maxwell. The electrical field E(t) generates a current density J(t) as a sum of conduction and displacement currents.

$$\nabla \times H(t) = J(t) = \sigma_0 E(t) + \frac{dD(t)}{dt}$$
(14)

 σ_0 is the volume conductivity of that material. Since polarization P(t) also contributes to the displacement current (13), one can write:

$$J(t) = \sigma_0 E(t) + \varepsilon_0 \varepsilon(\infty) \frac{dE(t)}{dt} + \frac{dP(t)}{dt}$$
(15)

Now, E(t) will be specified as a Heaviside step function H(t) of amplitude E_0 . For this, [Zaengl 2003] derives from (15) using a convolution in time domain the total current density J(t) as

$$J(t) = \sigma_0 E_0 + \varepsilon_0 E_0 [\varepsilon(\infty)\delta(t) + f(t)]$$
(16)

with
$$\mathcal{E}(\infty) = 1 + \chi(\infty)$$
 (17)

 $\varepsilon(\infty)$ is the high frequency part of the permittivity and $\delta(t)$ the delta-function as the derivative of the applied Heaviside step function H(t). f(t) indicates the "pure" dielectric response function, which illustrates the time dependence of the susceptibility or of the relative permittivity. f(t) monotonously decays in linear dielectrics.

$$f(t) = \frac{d\chi(t)}{dt} = \frac{d\varepsilon_r(t)}{dt}$$
(18)

Equation (16) is worthy of a closer look. The first term represents a *constant* resistive current caused by the DC conductivity σ_0 of a material. The second term stands for *fast* polarization processes resulting in a delta function $\delta(t)$ when a Heaviside step voltage is applied. Finally the third term describes the slow polarization processes, that are - together with conductivity - of interest for the detection of moisture in the context of this work.

Measurement by Polarization and Depolarization Currents

With a charging voltage of magnitude U_C applied at time $t = t_0$ and a geometric capacitance C_0 a polarization current $I_{pol}(t)$ can be defined [Zaengl 2003].

$$I_{pol}(t) = C_0 U_0 \left[\frac{\sigma_0}{\varepsilon_0} + \varepsilon(\infty) \delta(t) + f(t) \right]$$
(19)

In practical cases the middle part with $\varepsilon(\infty)$ cannot be measured because of its very short duration and broad dynamic range [Houhanessian 1998].

If a short circuit is applied on the measurement object after the charging time t_c the depolarisation current $I_{dep}(t)$ can be recorded.

$$I_{dep}(t) = -C_0 U_0 [f(t) - f(t + t_C)]$$
(20)

The very short discharging current pulse depending on $C_o \cdot \varepsilon(\infty)$ cannot be measured and is not included in (20). The term with t_C can be neglected for long charging times t_C , hence the depolarization current $I_{dep}(t)$ becomes equivalent to the dielectric response function [Houhanessian 1998].

DC Conductivity

Polarisation processes are superimposed on a time-independent DC conductivity σ_0 . It represents the movement of free charge carriers in cellulose and in oil. Conductivity could be determined by the DC component of the polarisation current. Actually this needs a very long time of some days at ambient temperature because of the superimposed slowly decaying polarisation effects. More useful is a calculation using the difference between polarisation and depolarisation currents.

$$\sigma_0(t_{\max}) = \frac{\varepsilon_0}{C_0 U_0} \left[I_{pol}(t_{\max}) + I_{dep}(t_{\max}) \right]$$
(21)

 t_{max} denotes here the polarisation and depolarisation current values for the longest measurement time available. Using a longer time increases the accuracy.

The specific properties of conductivity in oil and pressboard are further explained in the sections 4.3 and 4.4.

Superposition of Polarisation and Conductivity at an Oil-Paper-Insulation

A measurement at the dielectric of a real transformer always means to measure a superposition of various effects. Firstly these are the properties of the single materials as polarisation and conductivity of cellulose and conductivity of oil. Secondly the multilayer transformer insulation causes interfacial polarisation. Thirdly the water content and conductive aging products influence the dielectric properties, especially the losses. Finally temperature greatly increase the measured currents and shifts their shape to the left in Figure 44. The qualitative illustrations in Figure 44 are drawn supposing double-logarithmic axes. The figure illustrates that a clear separation between the different influences constitutes the key point for moisture determination.



4.2.3 Dielectric Properties in Frequency Domain

Current Density

To transform the equations from time to frequency domain, the material is assumed to be isotropic and linear. Under these conditions equivalent information can be obtained during a measurement in time or frequency domain.

With equations (11) and (15), the frequency domain representation of (16) is:

$$J(\omega) = \sigma_0 E(\omega) + j\omega\varepsilon_0 E(\omega)[1 + F(\omega)]$$
⁽²²⁾

 $(\mathbf{n}\mathbf{n})$

1 is the Fourier transform of the delta function and $F(\omega)$ the dielectric response function in frequency domain [Zaengl 2003]. The dielectric response can be calculated by the Fourier transform of f(t).

$$\underline{\chi}(\omega) = \underline{F}(\omega) = \chi'(\omega) - j\chi''(\omega) = \int_{0}^{\infty} f(t)e^{-j\omega t}dt$$
(23)

The resultant total current density in frequency domain can be expressed by combining (22 and (23).

$$\underline{J}(\omega) = j\omega\varepsilon_0 \left\{ \varepsilon(\infty) + \chi'(\omega) - j \left[\frac{\sigma_0}{\varepsilon_0 \omega} + \chi''(\omega) \right] \right\} \underline{\underline{E}}(\omega)$$
(24)

The imaginary part of the current density in (24) represents its *capacitive* component. The real part includes a resistive current due to the DC conductivity σ_0 and a resistive current due to dielectric losses χ ". The inertia of the dipoles and charge carriers moved by the electric field cause these dielectric losses. In addition, the dielectric permittivity consists of a real and an imaginary part, as the following equation depicts.

$$\underline{\varepsilon}(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega) = \left[1 + \chi'(\omega) - j\chi''(\omega)\right]$$
(25)

Thus the current density in (24) can be expressed shorter:

$$\underline{J}(\omega) = j\omega\varepsilon_0 \left[\varepsilon'(\omega) - j\varepsilon''(\omega)\right] E(\omega)$$
(26)

Measurement in Frequency Domain

With application of a sinusoidal voltage $U(\omega)$ and a capacitance $C(\omega)$ a sinusoidal current $I(\omega)$ will flow.

$$\underline{I}(\omega) = j\omega \underline{C}(\omega) \underline{U}(\omega) \tag{27}$$

With (24) this can be written in more detail as:

$$\underline{I}(\omega) = j\omega C_0 \left\{ \varepsilon(\infty) + \chi'(\omega) - j \left[\frac{\sigma_0}{\varepsilon_0 \omega} + \chi''(\omega) \right] \right\} \underline{U}(\omega)$$
(28)

$$\underline{I}(\omega) = j\omega C_0 \underline{U}(\omega) [\varepsilon'(\omega) - j\varepsilon''(\omega)]$$
⁽²⁹⁾

The capacitance is now a complex number consisting of losses $C''(\omega)$ and "pure" capacitance $C'(\omega)$.
$$\underline{C}(\omega) = C'(\omega) - jC''(\omega) = C_0 [\varepsilon'(\omega) - j\varepsilon''(\omega)]$$
(30)

$$C'(\omega) = C_0 \left[\varepsilon(\infty) + \chi'(\omega) \right]$$
(31)

$$C''(\omega) = C_0 \left[\frac{\sigma_0}{\varepsilon_0 \omega} + \chi''(\omega) \right]$$
(32)

The loss capacitance $C''(\omega)$ in (32) includes resistive and dielectric losses. A measuring instrument will always indicate both quantities and a discrimination is not possible. Since the measured permittivity $\varepsilon_{r,m}$ includes the conductive losses as well, it is different from the ideal permittivity ε_r as defined in (12).

The well-known dielectric dissipation factor $\tan \delta$ is the quotient of the loss part and the "pure" capacitive part of $C(\omega)$.

$$\tan \delta(\omega) = \frac{C''(\omega)}{C'(\omega)} = \frac{\varepsilon''(\omega)}{\varepsilon'(\omega)} = \frac{\frac{\sigma_0}{\varepsilon_0 \omega} + \chi''(\omega)}{\varepsilon(\infty) + \chi'(\omega)}$$
(33)

Equation (33) indicates the frequency dependence of the dissipation factor tan δ . Thus a measurement at one single frequency won't give comprehensive information about the insulation constitution.

The real part of permittivity $\varepsilon'(\omega)$ is here split into a constant high frequency part $\varepsilon(\infty)$ and a frequency-dependent susceptibility $\chi'(\omega)$. In reality, also $\varepsilon(\infty)$ is a function of frequency, but not within the frequency range considered here of up to some kHz.

Influence of Temperature on Dielectric Properties

Temperature influences intensely the dielectric properties as it changes the polarisation processes and the conductivity of a material. If depicted in frequency domain, the influence results in a shift of the measured spectral function along the frequency axis. The shape of the curve is thereby less influenced. Thus it is possible, to introduce a so called "master curve", that determines the behaviour of one material by shifting it along the frequency axis at various temperatures, [Du 2002], [Ekanayake 2006].

The logarithmic shift of the spectral function from temperature T_1 to T_2 expresses an Arrhenius equation, actually a formula for the temperature dependence of a chemical reaction rate. So the shift from one frequency ω_1 to another ω_2 can be calculated by:

$$\log(\omega_1) - \log(\omega_2) = \frac{E_a}{k} \left(\frac{1}{T_2} - \frac{1}{T_1} \right)$$
(34)

 E_a is the activation energy of the material, which can be determined by two measurements at different temperatures. k is the Boltzmann constant with $k = 8,617385 \cdot 10^{-5} \text{ eV} / \text{K}.$

The following equation compensates the conductivity for a temperature change from T_1 to T_2 . For example it can be applied for mineral oil to compare measurements at different temperatures.

$$\sigma_2 = \sigma_1 e^{\frac{E_a}{k} \left(\frac{1}{T_1} - \frac{1}{T_2}\right)}$$
(35)

Superposition of the Dielectric Properties of Cellulose and Oil

While measuring the dielectric properties of an oil-paper-insulated power transformer, the dielectric properties of the single materials are superimposed together with interfacial polarisation. The upper row of Figure 45 displays the dissipation factor curves of cellulose and oil with double-logarithmic axes.



Figure 45: Superposition of dielectric properties: Single impacts of the materials (upper row) and superposition at a multilayer insulation with influences of interfacial polarisation, oil conductivity, water and temperature (lower row)

Together with interfacial polarisation, the dielectric response shows a typical sshaped curve when (left drawing in lower row of Figure 45). The very low and the high frequencies reflect the cellulose, the steep slope the oil and the local maximum the interfacial polarisation effect, i.e. the insulation geometry. To a certain extend, the insulation geometry will also influence the areas dominated by cellulose. Water content and oil conductivity shift the dielectric response towards higher frequencies; however the shape remains similar (middle drawing in lower row of Figure 45). Temperature further increases the losses of cellulose and the oil conductivity (right drawing in lower row of Figure 45). Since moisture especially affects the low frequencies, data on the left hand side of the area influenced by the interfacial polarisation effect are required for a reliable moisture determination. Because the traces for different moisture content vary only in the low frequency area (Figure 64), it is not sufficient to measure the high frequency area only.

4.2.4 Modelling of Dielectric Properties

Representation by the XY-Model

The XY-model represents the volumetric fraction of two materials in one insulation system. In the cylinder-shaped transformer insulation all pressboard barriers and insulation paper are made into one single barrier with relative thickness X. All spacers of the cylindrical insulation form one with width Y. All the oil ducts form one oil duct of relative thickness 1-X and width 1-Y, Figure 46. This idea was at first used to model oil-paper-insulated cables, for example in [Kuechler 1996].



Figure 46: Representation of a cylindrical transformer insulation by the XY-model

Based on this aggregation the total permittivity $\varepsilon_{tot}(\omega)$ of a multilayer insulation can be calculated by the following equation.

$$\underline{\varepsilon}_{tot}(\omega) = Y \cdot \underline{\varepsilon}_{PB}(\omega) + \frac{1 - Y}{\frac{1 - X}{\underline{\varepsilon}_{Oil}(\omega)} + \frac{X}{\underline{\varepsilon}_{PB}(\omega)}}$$
(36)

Suppositions of this model are that the dielectric properties of pressboard in barriers and in spacers are identical and linear.

 $\varepsilon_{PB}(\omega)$ is a complex, frequency dependent vector. $\varepsilon_{Oil}(\omega)$ is the permittivity of oil. Since oil has only a small polarisability, the following equation covers its dielectric properties.

$$\underline{\varepsilon}_{Oil}(\omega) = 2, 2 - j \frac{\sigma_{Oil}}{\varepsilon_0 \omega}$$
(37)

The real part of $\varepsilon_{Oil}(\omega)$ as 2,2 is similar for all mineral oils, but much higher for vegetable oils. A closer look at the properties of oil conductivity σ_{Oil} gives section 4.3. There a frequency or time dependent conductivity $\sigma_{Oil}(\omega)$ is introduced, which describes the behaviour of oil more exactly. However, equation (37) is satisfactory for a first approach.

Analogue by an Equivalent Network

A descriptive way to model the dielectric properties are networks of resistors and capacitors. These RC-networks emulate the behavior of a real oil-paperinsulation. Prepositions are a monotonously decaying response function and a linear behavior of the dielectric.



Figure 47: Modelling of the dielectric properties of a multilayer insulation by an equivalent circuit

Figure 47 depicts the equivalent circuit for a multilayer insulation of oil and pressboard. Pressboard is characterised by a capacitance, a resistance and a several RC series connections. For oil, description by a capacitance and a resistance is sufficient because of its small polarizability. The values of the unknown elements in Figure 47 may determine a modelling procedure as described in [Houhanessian 1998]. This author uses a sequential algorithm and also bases the transformation between time and frequency domain on this model of equivalent circuits.

4.3 Dielectric Properties of Oil

This section investigates oil conductivity under the influences of measurement voltage and time, moisture, aging and temperature.

4.3.1 Ionic Conduction in Mineral Oil

Impurities give rise to the volume conductivity of mineral insulation oil, similar to solid dielectrics. Pure insulation oils, as saturated hydrocarbons, shows

neither ions nor free electrons. After refinery, some pollutants, moisture and acids remain providing ions as the dominating charge carrier in liquids.

Ions may arise from spontaneous dissociation of ionisable substances. The separation into ions under the influence of water is called electrolytic dissociation. Water alone has a conductivity of $\sigma = 6 \cdot 10^{-6}$ S/m at a temperature of 25°C because of the dissociation of H₂O to H⁺ and OH⁻. This results in a ion concentration of N_{H+} = N_{OH-} = 6,02 \cdot 10¹⁹ m⁻³ at 25°C [Münch 1987].

Water of course exists in mineral oils only in traces of some $\mu g/g$. Conductivity is intensified by other ionisable substances as for example carbon and carboxylic acids. Mineral oil also contains acids after refinery and especially because of oil and paper aging (p. 44). These acids dissociate as follows:

$$H-COOH + H_2O \rightarrow H-COO^- + H_3O^+$$
(38)

H-CO is the aldehyde group of a carboxylic acid, which is for example CH₃CO as acetic acid. The ion concentration of acetic acid comes up to $N_{H^+} = N_{Ac^-} = 6 \cdot 10^{23} \text{ m}^{-3}$ at 25°C already at a molar concentration of 0,1.

In particular the lower molecular acids are responsible for the increased conductivity. This is similar to moisture solubility and breakdown voltage, where these stronger acids have the more intense impact.

Beside organic acids, carbon as a product of cellulose deterioration, bases and salts also contribute to electric conductivity. [Link 1966] mentions additionally macro molecules from polymerisation and polycondensation of hydrocarbons.



Ions differ in the velocity of their movement. Therefore current and conductivity depend on time or frequency. According to [Link 1966] fast polarisation processes cause area A in Figure 48. B is dominated by fast ions. In C space charges increase the volume resistance. Continually dissociating ions then cause area D. From the figure it becomes obvious that the application of an electric field results in an "electrical cleaning" effect.

4.3.2 Setup and Samples

Measurement Setup

The dielectric properties of mineral oil were measured in time and frequency domain. Measurements in dielectric liquids are very sensitive to disturbances. A guarded measurement cell was built derived from [IEC 247]. The gap between the cylindrical electrodes is 2 mm. The capacitance in air C_0 is 59,5 pF, obtained from calculations and verified by measurements.



For the measurements in time domain an electrometer Keithley 6517A with built-in voltage source up to 1 kV was used [Keithley 1996]. This instrument covers a current range from 1 fA to 20 mA. The data was automatically logged by a software program. Keithley specifies the accuracy of its electrometer in the 2 nA range with ± 0.2 % of the reading + 30 counts and in the 200 pA range with ± 1 % of the reading + 5 counts. However the first 2,5 s in the pA-range are affected by the preamp settling times and are therefore uncertain.

The following equation derived from (19) calculates oil conductivity.

$$\sigma(t) = \frac{\varepsilon_0}{U_C C_0} i(t) \tag{39}$$

 U_C is the applied DC voltage, C_0 the empty capacitance of the measurement cell. The equations validity is restricted to materials with negligible polarizability.

An insulation diagnostic system IDA 200 made by Programma measured the oil properties in frequency domain [GE 2007]. The system measures the complex capacitance $C(\omega)$ and the dissipation factor $tan \,\delta(\omega)$ between 0,1 mHz and 1 kHz. Programma gives an accuracy for the capacitance measurement with IDA 200 within the range used here of ± 2 % of the reading + 1pF.

From the dissipation factor measurement the oil conductivity can be calculated with the following equation, derived from (33). A negligible polarizability and a real part of the permittivity of 2,2 are supposed.

$$\sigma(\omega) = 2, 2\varepsilon_0 \omega \tan \delta(\omega) \tag{40}$$

Investigated Oil Samples

Five mineral oils were investigated: new Shell Diala D with a neutralisation number of 0,03 mg KOH / g oil, new Nynas Nytro 3000 with a neutralisation number of 0,01 mg KOH / g oil, used Shell Diala D with a neutralisation number of 0,03 mg KOH / g oil (original filling of the Pancake model on p. 63), 25-years service aged oil with a neutralisation number of 0,08 mg KOH / g oil and service aged Shell K6 SX from 1965 with a neutralisation number of 0,49 mg KOH / g oil.

4.3.3 Effects of Field Strength, Moisture and Temperature

Conductivity and Polarizability of Mineral Oil

Figure 50 shows the polarisation and depolarisation currents for the service-aged Shell Diala D at a temperature of 21°C measured at a field strength of 70 V/mm.



This oil shows effects similar to Figure 48. Space charges increase the volume resistance at times longer than twenty seconds. The depolarisation current is 200 times less than the polarisation current. This justifies its neglect in the dielectric response model on p. 76.

Influence of Field Strength and Aging

Figure 51 displays the conductivity of insulation oil as a function of measurement time and field strength. New Nynas Nytro 3000 shows the impact of a cloud of charged ions as described in Figure 48. At the low voltage of 5 V/mm this happens at times longer than 30 s. For the service-aged Shell K6SX

this effect is nearly invisible. Here the fast ions dominate the ionic conduction up to 1000 s. In general, the charge cloud effect appeared especially in dry and new oils. With increasing moisture, aging and temperature the effect disappears.



Figure 51: Oil conductivity as a function of time and field strength for new Nynas Nytro 3000 (left) and service-aged Shell K6SX (right), measured at an oil temperature of 40°C and a relative saturation of 35 %

The theory of polarisation and conduction processes assumes a linear behaviour of dielectric materials (p. 69). The conductivity of oil clearly violates this condition, since it depends on time *and* on measurement field strength. On p. 103 an analysis approach is described that takes this behaviour into account.

The conductivities measured in the frequency domain agree well with the results in the time domain, as long as the space charge does not affect the measurements. The concurrent area comprises 1 s < t < 10 s in time domain and 1 Hz > f > 0,1 Hz in frequency domain, but varies with the oil and measurement conditions. This might be due to a limited validity of equation (40). It calculates oil conductivity from the dissipation factor, but is restricted to materials without polarisation effects. Yet the ionic space charge constitutes a kind of polarisation.

Oil aging results in increasing acidity. Figure 52 depicts the oil conductivity depending on the total acid number including a trend line. The figure includes the oils measured within this research work. The deviation from the trend line indicates, that the total acid number only roughly correlates with oil conductivity. A clearer dependence might be given by a relation to stronger, low-molecular acids. On the other hand, the total acid number will not indicate other conductive substances such as carbon as a result of pyrolysis.



Figure 52: Oil conductivity depending on total acid number, additionally a trend line

Influence of Moisture

The moisture saturation in oil was varied from 3,6-46 %. Figure 53 shows the corresponding oil conductivity for new Nynas Nytro 3000 at 21°C. An increasing moisture saturation increases the oil conductivity as well. This agrees to the considerations about dissociation of carboxylic acids on page 78.



Influence of Oil Temperature

To compare the temperature influence on the different oils single points of oil conductivity were used instead of the whole time range. These points are 10 s in time domain and 0,1 Hz in frequency domain. The time domain data agree well with that from frequency domain, however in most cases the frequency domain conductivity somewhat undercuts that of time domain. Figure 54 shows the results for new Shell Diala D, new Nynas Nytro 3000, service aged for 25 years and service aged Shell K6SX. Because of the construction of the measurement cell the relative saturation could not be held constant and it decreased from 35 % at 20°C to 5 % at 80°C.



The activation energy of the specific oil causes the different gradients in Figure 54. Activation energy characterizes the temperature dependence of a material property, usually for chemical reactions, but here for conductivity. The following table gives the activation energies of the four mentioned oils and decides between the sources time domain and frequency domain measurement. The activation energy was calculated by equation (35). Except for new Shell Diala D the results from the different measurement instruments agree.

Oil	E _a from time domain	E _a from frequency domain
Diala D	0,43	0,35
Nytro 3000	0,54	0,52
25a aged	0,32	0,37
Aged K6SX	0,32	0,33

Table 6: Activation energy E_a of four investigated oils

Accuracy of the Results

The accuracy of the oil conductivity measurements depends on the measurement cell and the instruments.

Since the measurement cell is not sealed, it was difficult to keep the relative moisture saturation stable. So the curves in Figure 53 are subject to this systematic error. It typically affected the low moisture results, where the moisture saturation increased quickly. For the measurements at increasing temperature (20-40-60-80°C) the moisture saturation decreased correspondingly (approximately 35-15-10-5 %). Thus the conductivity results in Figure 54 were 2-3 times higher at constant moisture saturation. This also affects the activation energy in Table 6, which would be 20-40 % higher.

The formation of a space charge made by ions in the measurement cell further complicates the measurements. The space charge appears especially for dry or

low conducting oils and can be interpreted as a polarization effect. However, the calculation of oil conductivity according to (39) and (40) requires the absence of polarization effects. This space charge especially impairs the measurements at longer times or at low conductivities.

Compared to the two mentioned sources of uncertainty the inaccuracy of the measurement instruments plays a minor role.

The results will also not be influenced by matter transport at ionic conduction because of the low ion concentration.

4.4 Dielectric Properties of Cellulose Models in Time Domain

This section investigates the dielectric properties of pressboard in time domain under the influences of moisture content and temperature. Special attention was given to the linearity of the dielectric response.

4.4.1 Investigated Samples and Measurement Setup

The influences of moisture content, insulation temperature and measurement voltage were investigated at pressboard and paper samples. The disc-shaped samples had a diameter of 160 mm and a thickness of 1,5 mm for pressboard and 50-60 μ m for paper. The moisture content relative to weight for the new cellulose samples were 0,6; 1; 2; 3 and 4 %. To obtain the desired moisture content, the samples were first dried in a vacuum oven and then exposed to the atmosphere while a scale measured their weight. After reaching the desired weight because of water adsorption from air the samples were oil impregnated.

New transformer oil Nynas Nytro 10 GBN served for cellulose impregnation and filled the measurement cell. Thermal treatment at 130°C for 3 months aged samples with 0,6 and 4 % moisture content immersed in oil. The degree of depolymerisation of the aged samples with high water content decreased from 1000-1400 to 200-400. The thermal aging process altered the water content to 0,9 and 2,8 %.

It is very important to mention, that the within this section 4.4 given values for moisture relative to weight were not obtained by the Karl Fischer titration procedure as favoured earlier in this work (chapter 1, appendix 7.1). The heating temperature for cellulose samples was 135°C instead of 160°C. Thus the procedure favoured in this work would reach moisture contents around 15 % higher then the ones given in this section 4.4. The samples for the time domain

measurement in section 4.4 prepared the Poznan University of Technology within the European research project $REDIATOOL^2$.

Measurement Setup

The electrometer Keithley 6517A, as introduced at p. 80, served for the measurement of polarisation and depolarisation currents of cellulose samples. During all measurements, a voltage of 10 V drove the currents. With the 1,5 mm pressboard samples this results into a field strength of 6,7 V/mm. This low value prevented nonlinear behaviour. For automated measurements control and data acquisition software was written.

A guarded measurement cell prevented disturbances by parallel current paths and electrostatic fields. The plain electrodes were made of aluminium and the cell walls of acrylic glass. The sense electrode had a diameter of 113 mm, resulting in a geometric capacitance C_0 of 59,2 pF for the 1,5 mm pressboard samples and 250 pF for five paper sheets. To ensure good contact of the electrodes, a weight of 5 kg was added to the upper electrode. The measurement cell was additionally shielded by a climatic chamber. A personal computer recorded the measurement data from the electrometer (Figure 55).



Figure 55: Measurement setup for the time domain measurements

4.4.2 Influences of Moisture, Temperature, Aging and Geometry

Representation of the Results

Measurement data should be displayed in a way that allows for an easy comparison to other data. The representation in *frequency domain* by the permittivity $\varepsilon(\omega)$ ensures good comparability. In *time domain* the presentation by i(t) is still common. Unfortunately here also characteristics of the measurement setup such as voltage and dimension of the test object also

² European research project REDIATOOL: "Reliable Diagnostics of HV Transformer Insulation for Safety Assurance of Power Transmission System", contract NNE5/2001/472

influence the representation. These are U_0 and C_0 in equation (19). To display solely the properties of the insulation material, the measured currents were therefore normalized by relating them to the measurement voltage U_0 and geometric capacitance C_0 . The following equation thereby illustrates the received normalized currents I_{pn} . The unit is then 1/s.

$$I_{pn} = \frac{I_{pol}(t)}{C_0 U_0} = \frac{\sigma_0}{\varepsilon_0} + f(t)$$
(41)

Influence of Moisture Content

Figure 56 shows the polarisation and depolarisation currents under the influence of moisture in pressboard at an insulation temperature of 23°C.



Figure 56: Normalized polarisation (left) and depolarisation (right) currents at 23°C for 0,6; 1,0; 2,0; 3,0 and 4,0 % moisture content. The depolarisation currents disappear in noise below 0,02 1/s.

A clear dependence on moisture content is visible, the amplitude increases with moisture and the currents reach their final value earlier. Depolarisation currents show the same initial amplitude as polarisation currents. They decrease faster for samples with higher moisture content. The electrometers settling time of 2,5 s causes errors during the first few seconds of the measurement.

Influence of Insulation Temperature

Figure 57 illustrates the strong influence of temperature on the insulation properties at a moisture content of 4 %. The conductivity of pressboard and oil increases significantly with temperature and the current amplitudes increase as well. Polarisation processes occur much faster and the final value as an equivalent for pressboard resistance and moisture content in paper is reached earlier with increasing temperature.



Figure 57: Polarisation currents at 23, 40, 60 and 80°C for 4 % moisture content

Power transformers are operated at elevated temperatures of 30-80°C middle oil temperature. Because of the intense influence of insulation temperature a reliable temperature compensation is essentially needed to interpret measurement data. Since distinguishing between the influence of moisture and of insulation temperature is hardly possible, a reliable temperature measurement is of utmost importance. Measuring the temperature directly in a transformer oil sample usually gives more dependable results than using the built-in thermometer of a transformer.

Pressboard versus Paper

Figure 58 illustrates the differences between pressboard and Kraft paper with the same moisture content of 3 %. The end values of the polarisation currents, reflecting resistance, are the same. The differences in the shape of the curve (up to 30 s) are due to interfacial polarisation processes that have a broader influence on the five layers of paper sheets. Furthermore this measurement proves the validity of the XY-model, which combines paper and barriers of the insulation into one single barrier.



Figure 58: Polarisation and depolarisation currents of pressboard and paper having a moisture content of 3 % at temperature of 23°C

Influence of the Aging State

Figure 59 compares polarisation and depolarisation currents for new and aged pressboard samples. The initial amplitude and the end values are higher for aged materials. Conductive aging by-products in cellulose increase the losses and conductivity in addition to moisture. These aging by-products are e.g. carbonic acids originating from cellulose decomposition. [Houhanessian 1998] also reported this behaviour.



Figure 59: Comparison of PDC for new and aged pressboard with 4 % moisture content at 23°C

Influence of Geometry

Figure 60 depicts the influence of insulation geometry on polarisation currents at 23°C. One model had a ratio of 20 % barriers to oil and the other 33 %. With an increasing portion of insulation oil (oil duct), the current amplitudes are higher. Because of the oil the conductivity of the model increases. Additionally the currents remain nearly constant for a longer time. Interfacial polarisation at the interfaces between oil and pressboard cause this effect.



Figure 60: Influence of insulation geometry on polarisation currents at 0,6 % moisture content

4.4.3 Linearity of the Dielectric Properties

It is common to describe polarization processes in dielectrics by a linear model of R-C-networks as originally published by [Debye 1934], (Figure 47). However some authors observed a nonlinear behavior of multilayered oil-paper-insulations [Kuechler 2004].



Figure 61: Nonlinear behaviour of oil-paperinsulations with multiple layers at a field strength of 100 V/mm with and without oil duct

Figure 61 shows a comparison of polarization currents at pressboard models with and without oil duct. To support the nonlinear behavior an extreme field strength of 100 V/mm was chosen. A ratio of "1" (red dashed line) stands for a linear behavior. This is more or less given for a pressboard insulation without an oil duct. Due to the dependence of oil conductivity on time and field strength the behavior of an insulation with oil duct is nonlinear. RC-networks according to Debye can not emulate such properties. The nonlinearities increase with field strength and are negligible only for less then 20 V/mm. Of course, the question arises, do these nonlinearities also appear in real power transformers. In this case a measurement voltage of $200 V_{DC}$ at a thickness of 50 mm of the main insulation would be typical values. The resulting field strength of 4 V/mm will not cause nonlinear behaviour due to the properties of oil conductivity.

Accuracy of the Results

The accuracy of the measured polarisation and depolarisation currents depends on the instrument accuracy, the measurement setup and special properties of the materials.

The instrument accuracy of the Keithley 6517A as described on p. 80 is an error source of minor importance except for the settling time of the input amplifiers. This affects the first 2,5 s of a measurement at current ranges of 200 pA or less.

The measurement setup implies an error due to oil layers at the surface of the pressboard samples. All the pressboard discs have a - sometimes very small - curvature so an oil layer interferes with the properties of pressboard. Although a weight of 5 kg tried to decrease this influence, an interfacial polarisation occurs and additionally the transient properties of oil conductivity affect the

measurement. This also diminishes the repeatability of the measurements, for a new setup the dimensions of the oil layer change. A thorough investigation of this influence gives p. 99 in the context of the frequency domain measurements.

The special properties of oil conductivity not only interfere because of the oil layers, but also by the oil in the macropores of the pressboard. Section 4.5.3 (p. 96) deals with the influence of the impregnating oil.

For the time domain measurements a so called "Battery Effect" causes a current which is superimposed onto the current from the voltage source. The current can be measured, even if no external field is or was applied. The "Battery Effect" appears only at elevated temperatures (60-80°C) and its amplitude is far beyond that of the measurement current. Hence under the conditions of this measurement setup the "Battery Effect" can be neglected.

As alluded to on p. 85, the moisture contents in cellulose mentioned within this section 4.4 were not obtained by the favoured procedure for moisture titration. To make it comparable to the other results of this thesis, around 15 % should be added to the given values.

4.5 Dielectric Properties of Cellulose Models in Frequency Domain

Moisture determination by algorithms analyzing dielectric properties depends strictly on the reliability of the database obtained from laboratory measurements. To get dependable data for the database requires careful measurements on pressboard and paper samples in the laboratory. The dielectric properties of new and aged pressboard were determined under various conditions. Emphasis was on the impact of aging on the dielectric properties. Moreover the influence of the type of pressboard and the oil used for impregnation were observed.

4.5.1 Investigated Samples and Measurement Setup

Treatment of New Pressboard Samples

Square shaped pressboard samples with a thickness of 2 mm, an edge length of 155 mm and 60 g weight served to investigate the influences of moisture content, temperature and aging. To obtain various water contents, at first an oven dried the samples at 150°C for several hours. After this the samples had a water content of 0,4-0,7 % relative to weight. Then the samples were closed in stainless-steel containers and small vessels with water put into the containers (Figure 62). Three periodical heating cycles of 3 h up to 90°C then evaporated the water from the small vessels, which then migrated into the cellulose samples. Cycles are needed to generate a homogenous moisture distribution. According to the mass of added water various moisture contents were obtained. For the treated samples the Karl Fischer titration measured water contents of 0,8; 2,0;

2,8; and 3,9 % following the procedure in section 7.1. Three pressboard samples for each moisture level were made.



Figure 62: Moisturization of pressboard samples in the oven by water vapour from separate vessels

Aging of the Pressboard Samples

For artificial aging, three samples of each moisture level were put into four stainless-steel containers. Metal shavings in paper bags aimed to emulate the conditions in real transformers: 5 g copper, 5 g iron, 1 g aluminium and 1 g zinc per container. Thermal energy aged the samples at 130°C for 1500 hours (2 months) in closed containers, that is in absence of oxygen. Hereafter the degree of polymerisation decreased to 110-150, independent of the original water content. After aging the water content in the pressboard samples was only 1,0-1,2 %. The water evaporated due to the high temperature and caused a high pressure in the containers which was assumed to be through the silicon sealing the water vapour left the containers. Thus all the containers had a moisture *saturation* in equilibrium to air at 130°C, that is a moisture *content* of around 1%. In order to obtain various moisture contents, the aged samples were again moistened resulting in moisture levels of 1,2; 2,1; 3,1 and 3,9 %.

The total acid number of the oil surrounding the pressboard samples increased from 0,01 to 0,06 mg after aging. One reason for the low acidity might be that aging occurred under the absence of oxygen in the air.

Insulation Oils Used for Impregnation

To investigate the influence of moisture, temperature and aging as described above a new naphthenic mineral oil Nynas Nytro 3000 was used.

A second setup aimed to find out the influence of oil conductivity on the dielectric properties of pressboard. Here the moisture content of all pressboard discs was left at two levels (0,7 and 2,6 %), but oils with different conductivities were used for impregnation. A new Nynas Nytro 3000X with 0,05 pS/m, a new TR8 with 3,5 pS/m, a service-aged oil with 21 pS/m and a service-aged Shell K6SX with 148 pS/m, measured at 21°C and at 10 s after voltage application (p. 83). The acidity of the oils were 0,05; 0,29; 0,3 and 0,3208 mg KOH/g oil.

Pressboard Samples of Various Manufacturers

Another question is, how does the pressboard material relate to the dielectric properties, and are there differences because of different manufacturers? Therefore four different pressboard samples were impregnated with Nynas Nytro 3000X: Weidmann Transformerboard III, Weidmann Transformerboard IV, Pucaro Elboard HD and a low density material from Petrocard. All samples had a thickness of 2 mm, a moisture content of 2,5 % and were measured at 23°C. Much care was taken to keep these parameters identical in order to have comparable measurement results. An important difference between the materials consisted in their density: 0,9 / 1,1-1,3 / 1,1-1,25 / 0,9 g/cm³.

Measurement Setup

The insulation diagnostic system IDA 200 [GE 2007] measured the complex capacitance $C(\omega)$ of pressboard in the frequency range 0,0001-1000 Hz. During all measurements a voltage of 10 V_{RMS} was applied. At the 2 mm pressboard samples this results in a field strength of 5 V/mm. This low value was designed to prevent nonlinear behaviour.

The same electrode arrangement as on p. 86 prevented disturbances by parallel current paths and electrostatic fields. Instead of walls made by acrylic glass here metallic ones were used. Weights of 10 kg at the upper electrode tried to ensure a good contact with the samples (Figure 63).





4.5.2 Results for Moisture, Temperature and Aging

Influence of Moisture Content

Figure 64 shows the permittivity as real part ε' and losses ε'' over frequency for four moisture contents: 0,8; 2,0; 2,8 and 3,9%. Since for each moisture level three samples were prepared, the moisture level in the individual samples varies slightly. Figure 64 displays the average value for each curve point. Water increases the losses because of its conductivity and since it enables acids to dissociate. Water also increases the polarizability, visible at ε' below 1 Hz, probably due to interfacial polarization in macro pores.



Figure 64: Influence of moisture on the dielectric properties of new pressboard at 20°C insulation temperature. The upper figure displays the real part of the permittivity, the lower figure the imaginary part (losses).







As presented in Figure 65, temperature greatly increases the losses and also the polarizability of pressboard below 1 Hz. Similarly to moisture content, the

thermal energy increases the conductivity of the material and also intensifies polarisation phenomena inside the material.

Influence of Aging

Some aging by-products such as acids are conductive; they dissociate with water and increase the losses. Thus aging causes a similar influence on the dielectric properties as water and temperature. Aging also changes the microstructure of pressboard. The increased polarizability ε' at frequencies below 10 Hz might be caused by interfacial polarisation in macro pores.



Figure 67 compares the dissipation factor of aged material to that of new material. For similar moisture content the losses in aged materials are much higher than they in the new material. Accordingly, a moisture analysis algorithm that does not compensate for conductive aging products will overestimate moisture content. This may lead to unnecessary drying out of transformers.



4.5.3 Influence of Oil Conductivity and Pressboard Material

The influence of oil conductivity on the dielectric properties of pressboard samples and that of the pressboard type are displayed in this section. Moisture analysis is based on a comparison of measured dielectric properties to modelled dielectric properties. The modelled dielectric properties are derived from laboratory measurements of pressboard samples. Thus the accuracy of the laboratory measurements determines that of the moisture analysis.

The laboratory measurements are usually done on impregnated pressboard discs. So how does the oil used for impregnation interfere with the measured dielectric properties? This will be answered in the following subsection. Another question is, are the dielectric properties of different pressboard types identical. To investigate this, the next but one subsection shows the results of low and high density materials from various manufacturers.

Influence of Oil Conductivity

Figure 68 depicts the influence of the conductivity of the oil used for impregnation of new pressboard samples on dissipation factor. Although the moisture content of all samples was identical with 2,6 %, the losses increased with the conductivity of the oil used for impregnation. Figure 69 shows the same behaviour for moisture in pressboard of 0,7 %.



The impregnation of new pressboard samples with high conductive oils results in similar dielectric properties as aging of pressboard and oil (Figure 67). Therefore it is proposed to use the knowledge of the oil conductivity for compensation of the influence of conductive aging by-products. This idea is further explained at p. 104.

Influence of Pressboard Material

Two high density and two low density pressboard samples made by three manufacturers were compared: Pucaro Elboard HD, Weidmann

Transformerboard IV, Weidmann Transformerboard III and a low density material from Petrocard.

Figure 70 shows the permittivity of the four pressboard samples. The high densitiy materials $(1,1-1,3 \text{ g/cm}^3)$ own a higher polarizability of $\varepsilon' = 4,6$ compared to the low density materials $(0,9 \text{ g/cm}^3)$ having $\varepsilon' = 3,0$. The high density materials simply contain more cellulose fibres per volume, thus more polarizable matter and therefore a higher polarizability. The losses, the imaginary part ε'' of permittivity, are higher for the high density materials as well.



The analysis software described on p. 101 analysed the measurements and gave moisture contents of 1,5; 1,6; 1,6 and 2,1 % for the four materials Weidmann TIII, Petrocard, Weidmann TIV and Pucaro HD. In reality the moisture content was identical. For the analysis of a real transformer the pressboard material is of course unknown. The different dielectric properties of pressboard materials limit the accuracy of moisture analysis methods to around $\pm 0,4$ % moisture by weight.

4.5.4 Accuracy of the Results

As for the time domain measurements, the accuracy depends on the instrument accuracy, the measurement setup and special properties of the materials.

The accuracy of the AC voltage and current measurement system IDA 200 as given at p. 80 is of minor importance compared to influences caused by the measurement setup.

A non-homogenous moisture distribution leads to a poor repeatability of the measurements. Therefore the moistening process was done in several heating cycles. After several cycles the water molecules migrate into the whole material forming a homogenous moisture distribution. Even a long equilibration time at ambient temperature doesn't give repeatable results, so several temperature cycles are necessary.

Influence of Oil Layers

The presence of oil layers between the electrodes and the pressboard sample causes a very important error influence. If even thin oil layers are present, an interfacial polarisation effect will overlay the dielectric properties of the sample. Heating of the pressboard discs often bends them slightly out of their original plain shape. Thus a thin oil layer exists, as illustrated in Figure 71. Especially for measurements at high temperature (80°C) the effect becomes dramatic, resulting into a poor repeatability.



To investigate this effect, a pressboard disc was measured immediately after moistening and two months later. For the first measurement the effect became visible as a "hump" in Figure 72, especially at the loss capacitance C". Then the sample was left for 2 months between the electrodes with a weight of 10 kg on the upper electrode. Figure 72 shows the dielectric properties before and after this pressing experiment, the "hump" disappeared. During this experiment the losses C" increased slightly, probably due to moisture ingress into the measurement cell.



Another experiment investigating the influence of oil layers was made with nonimpregnated pressboard. At first the dielectric properties were measured. Then with a conductive spray (copper particles) circular electrodes were sprayed on the pressboard disk and the measurement repeated. Figure 73 depicts the dissipation factor with and without the sprayed electrode. The sprayed electrode of course provides a perfect contact to the pressboard surface in contrast to the rigid plain electrodes. Because of the perfect connection the curves become smooth and the "hump" disappeared. Unfortunately the sprayed electrode is not regularly applicable, since the conductive liquid also ingresses into the material and therefore increases the losses immensely.



Figure 73: Dissipation factor of a nonimpregnated pressboard disc with rigid plain and with sprayed electrodes.

Software (p. 101) can also simulate this effect of a small oil gap. Figure 74 compares the dissipation factor of pressboard only to pressboard with a small oil layer. The software based on the XY-model (p. 77) calculated the typical

"hump" for a small oil gap of X = 90 % and Y = 20 % with an oil conductivity of 20 pS/m. For the measurements on 2 mm pressboard discs this means a thickness of the oil layer of 200 μ m, which can easily be reached.



Figure 74: Simulated dissipation factor of a pressboard disc with and without a thin oil layer

Because of the two experiments (pressed pressboard disc, sprayed electrodes) and the simulation it was decided to correct all results showing a "hump" in the middle of the measured frequency range. The correction was done by a comparison of the measured curves to ideal curves and to polynomial splines. For example, the curve with oil duct in Figure 74 would be corrected in the frequency range 2-0,01 Hz. All the measurements in frequency domain (section 4.5) having such a "hump" were corrected in this way.

It must be admitted, that the "hump" may also originate from specific properties of the dielectric as [Du 2002] and [Ekanayake 2006] assume.

4.6 Improved Moisture Analysis

This section describes approaches to improve moisture analysis based on the measurement of dielectric properties. It introduces a moisture analysis algorithm and considers the influence of conductive aging by-products.

Analysis Algorithm

Moisture analysis is based on a comparison of dielectric properties measured in a real transformer and modeled dielectric properties derived from a data base.

- 1. At first the capacitance $C(\omega)$ of the transformer with the unknown water content is measured down to low frequencies.
- 2. The insulation temperature T from the measured capacitance vector $C(\omega)$ is taken and the corresponding permittivity record $\varepsilon_{PB}(\omega)$ from the extraand interpolated data base. The data base consists of measurements of new pressboard as described at p. 93 ff.

- 3. Equation (36) of the so called XY-model combines this permittivity record $\varepsilon_{PB}(\omega)$ with the complex oil permittivity $\varepsilon_{Oil}(\omega)$ from equation (37). The XY-model allows for the computation of dielectric properties of a linear multi-layer-dielectric, where X represents the ratio of barriers to oil and Y the ratio of spacers to oil (p. 77). It is not necessary to enter the exact percentage values of the insulation geometry as area dominated by pressboard only on the low frequency side of the area dominated by the insulation geometry is available (Figure 45).
- 4. The obtained modelled permittivity $\varepsilon_m(\omega) = \varepsilon_{tot}(\omega)$ is then converted into a modelled capacitance $C_m(\omega)$ by multiplying an estimated geometrical capacitance C_0 .
- 5. Then the major part of the analyzing procedure takes place: the measured capacitance $C(\omega)$ is compared to the modeled capacitance $C_m(\omega)$. A fitting algorithm changes properties moisture, oil conductivity, and insulation geometry of the model, until it reaches the smallest error between both data, which means the best conformance between model and measurement.
- 6. The modeled capacitance $C_m(\omega)$ with the best agreement to the measured capacitance $C(\omega)$ gives the moisture content in cellulose and the oil conductivity of the real transformer.



Figure 75 depicts the programming flowchart of the analysis algorithm. The algorithm was implemented in $Matlab^3$.

³ Numerical computing environment and programming language, created by The MathWorks Inc.

Fitting Error Detection

The error detection is based on a minimal modulus of the difference of dissipation factor curves. This gave the best results for transformers with a known moisture content. Figure 76 illustrates the deviation (error) between measurement and model after the fitting procedure.



Weighting of Low Frequency Data

Figure 64 depicts the connection between losses and frequency range; water increases the dielectric losses below 1 Hz. Thus the fitting algorithm particularly weights the low frequency data. By weighting the low frequency data the sensitivity of water analysis to a varied oil conductivity decreases substantially since oil conductivity especially influences the middle frequencies.

Transient Oil Conductivity

Since the conductivity of oil decreases with time (p. 81 ff), an approach was made to consider this phenomenon in the analysis software.

To implement this property into the analysis software, vectors of oil conductivity $\sigma(\omega)$ were used instead of single complex numbers according to equation (37). Unfortunately this did not improve the moisture analysis results, but on the contrary the results got worse. One possible explanation is the field strength dependence of oil conductivity. During on-site measurements the field strength might be much lower than in the oil test cell. Supposing a test voltage of 200 V and an insulation thickness of 50 mm the field strength will be around 4 V/mm, thus the oil conductivity behaves in non-transient manner. Beside this for measurements on real transformers it is hardly possible to know the transient properties of the specific insulation oil.

Extended XY-Model

The original XY-model was extended to take into account the influence of parallel current paths. Parallel current paths provide a direct connection through

oil between conductors. Their influence seems to be extensive, since oil conductivity exceeds that of cellulose up to a thousand fold [Kuechler 2004]. To take account of these influences the original XY-model was extended by the geometrical parameter Z as depicted in Figure 77.





Equation (42) calculates the permittivity of the model.

$$\underline{\varepsilon}_{tot}(\omega) = \frac{1 - Y - Z}{\frac{1 - X}{\underline{\varepsilon}_{Oil}(\omega)} + \frac{X}{\underline{\varepsilon}_{PB}(\omega)}} + Y \cdot \underline{\varepsilon}_{PB}(\omega) + Z \cdot \underline{\varepsilon}_{Oil}(\omega)$$
(42)

This approach especially improves the analysis results for measurements with a ratio of pressboard to oil of more than 50 %. Below 50 % the influence of parallel current paths vanishes because of the large oil volume which is already implicit in the analysis. The main insulation of power transformers has a ratio of pressboard to oil of 17-50 %, thus parallel current paths might be neglected in practical cases. Additionally, the conductors of HV windings are usually wrapped in insulation paper.

Consideration of Conductive Aging By-products

Conductive aging products increase the losses in the dielectric in a very similar manner to water. Figure 67 displays the dissipation factor for new and aged pressboard in comparison. It impressively shows that, without compensating for conductive aging products, the analysis software will fail to accurately determine the water content in cellulose. This may initiate inappropriate maintenance actions such as drying.

A compensation approach was developed based on the conductivity of the insulation system. For conductive aging products a thermodynamic equilibrium exists similar to that for water. Here the chemical aspect plays the major role resulting in an equivalent effect of aging by-products on the dielectric properties in cellulose and oil. Based on this assumption a software approach to compensate for conductive aging products was developed. The approach uses the oil conductivity delivered by the fitting procedure (p. 101) to compensate for the effect on pressboard. Its mode of action displays an exemplary measurement on an aged transformer at p. 114.

Graphical User Interface

Figure 78 shows the graphical user interface of the developed Matlab software. The data display can show the permittivity, capacitance or dissipation factor. As model parameters the temperature and ratios of barriers to oil X and spacers to oil Y have to be entered. If unknown, the user can enter an estimated range, here 15-25 % for Y. Weighting enables an emphasis on special frequency ranges. The "Fitting" button starts the automatic fitting based on differences of dissipation factor. The result for oil properties delivers a calculated conductivity. Here also a conductivity measured at the corresponding temperature can be entered as well as the permittivity, which is different to 2,2 in, for example vegetable oils. The result for pressboard properties gives the moisture content in %, derived from the model. Further it displays the moisture saturation. This value is calculated from the moisture content and the insulation temperature via an analytical approximation of the moisture isotherm in Figure 20. It could also be directly derived from the data base, provided that each permittivity record is combined not only with water content, but also with water saturation. Rather experimental are the given values for breakdown voltage and bubble temperature. They are derived from analytical approximations of Figure 2 and Figure 3.



Figure 78: Graphical user interface of the developed software

Verification at the Pancake Model

The new software was verified at the comparative test, that the three commercialized dielectric diagnostic methods also passed. At p. 63 the commercialized techniques Recovery Voltage Method, Polarisation and Depolarisation Currents and Frequency Domain Spectroscopy were

benchmarked under three parameters: insulation geometry, insulation temperature and oil conductivity. As for the commercially available software the new one should be able to compensate for these parameters.

Figure 79 proves the advantages of the newly developed software in comparison to the analysis results of the commercial software as depicted in Figure 41, Figure 42 and Figure 43. The compensation for different oil conductivities works especially well because the low frequency data is weighted, as is the compensation for insulation geometry. Thus the error influence because of oil exchanges at real transformers is reduced. For increasing temperatures the analysed moisture content apparently decreases. This might be due to different temperature behaviour of the pressboard material in the model compared to that in the data base. Altogether the new software shows the smallest sensitivity to the investigated influences or in other words the best capability to compensate for them.



Figure 79: Moisture analysis results at the pancake model obtained with the new software. The dotted line marks the result of a Karl Fischer titration on paper samples taken out of the model.

5 On-Site Measurements at Power Transformers

"Tests need to be focussed on individual attributes in an attempt to arrive at an unambiguous diagnosis." [Guuinic 2001]

This chapter applies the described methods for on-site moisture assessment at real transformers. It confirms the findings of chapter 3 and chapter 4 in practical cases and focuses on a comparison of the obtained results to that of already established methods. This chapter starts with an overview of on-site measurements and then discusses the assessment of selected transformers. The issue of moisture non-equilibrium during a dielectric measurement is also taken into account.

5.1 Overview of Measurements

The following table gives an overview of the measurements on real power transformers made within this research work. It specifies the particular transformer and the measurement methods and compares the results in a bar graph. The "transformer" row displays the year of manufacture, rated power, turn ratio, vector group, cooling mode and oil temperature by the built-in thermometer.

The row "sampling" shows the results of moisture content in oil (ppm), moisture saturation of oil (%), oil sample temperature and moisture content of paper (%). It also indicates, whether the Karl Fischer titration was made at the Universität Stuttgart (W_{IEH}) or by another laboratory (W_L). The column also displays oil conductivity, if measured in the oil sample.

The row "Dielectric diagnostic" names the applied instruments, measurement parameters, the analysis method used to get moisture content and the modelled oil conductivity. Here FD means frequency domain and TD time domain.

The abbreviations in the bargraph column mean: Mods – moisture content as analysed by Programma MODS 1.5 [GE 2007], Dira – analysed by the software developed within this work (p. 101), RVM – analysed by Haefely SWRVM 2 V.3.0 [Haefely 2007], PDC – unless otherwise stated analysed by an own data set of PDC data, KF-I – sampling and subsequent Karl Fischer titration done by the IEH laboratory, KF-L – sampling and subsequent Karl Fischer titration done by an external laboratory, PPM – oil sampling and application of equilibrium diagram by [Oommen 1984] and RS – oil sampling and application of an moisture isotherm as proposed on page 57.

Transformer 1: Sampling:	2004, 400 MVA, 400/132/11 kV, YN/aO/d1, 21°C, autotransformer	5 4 3
Dielectric diagnostic:	FD: IDA 200, 140 V, 1000-0,0001 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0,06 \text{ pS/m}$	2 1 0,6 Mods
Transformer 2:	2004, 400 MVA, 400/132/11 kV, YN/aO/d1, 21°C, autotransformer	5 4
Sampling:	Not applied	3
Dielectric diagnostic:	FD: IDA 200, 140 V, 1000-0,00046 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0,21$ pS/m	2 1 0,5 Mods
Transformer 3:	2004, 400 MVA, 400/132/11 kV, YN/aO/d1, 21°C, autotransformer	5 4
Sampling:	Not applied	3
Dielectric diagnostic:	FD: IDA 200, 140 V, 1000-0,00046 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0,03$ pS/m	2 1 0,4 Mods 0
Transformer 4:	2003, 133 MVA, 420/10,5 kV, Ii0, OFWF, 27 / 32°C before and after heat run test, single phase transformer	5 4 3
Sampling:	Not applied	$2 - \frac{1}{08} - \frac{1}{$
Dielectric diagnostic:	FD: IDA 200, 140 V, 1000-0,001 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0,11$ /0,13 pS/m, TD: RVM 5462, t _C up to 2000 s	1 0,4 0,4 0,4 0,0 Mods RVM
Transformer 5:	1979, 800 MVA, 415/21 kV, YNd5, OFWF, 16°C, generator step up unit, stored as backup	5 <u>4,5</u> 4 <u>3</u>
Sampling.		
Sumpring.	$3,5 \text{ ppm at } 16^{\circ}\text{C}, \text{RS}_{\text{Oil}} = 1,3 \% \text{ at } 16^{\circ}\text{C}$	2 1,3 1,4 1,3
Dielectric diagnostic:	3,5 ppm at 16°C, $RS_{Oil} = 1,3$ % at 16°C FD: IDA 200, 140 V, 1000-0,00046 Hz, analysed by MODS 1.5 FD-TD combined: DIRANA 1, $\sigma_{mod} = 0,2$ pS/m	2 1,3 1,4 1,3 1 0 Mods Dira PPM RS
Dielectric diagnostic: Transformer 6:	3,5 ppm at 16°C, $RS_{Oil} = 1,3$ % at 16°C FD: IDA 200, 140 V, 1000-0,00046 Hz, analysed by MODS 1.5 FD-TD combined: DIRANA 1, $\sigma_{mod} = 0,2$ pS/m 1975, 266 MVA, 420/21/21 kV, Ii0, OFWF, 25°C, single phase transformer, scheduled for scrapping	2 1,3 1,4 1,3 1 0 Mods Dira PPM RS 5 7,0 4 3,0 7,0
Dielectric diagnostic: Transformer 6: Sampling:	3,5 ppm at 16°C, $RS_{Oil} = 1,3$ % at 16°C FD: IDA 200, 140 V, 1000-0,00046 Hz, analysed by MODS 1.5 FD-TD combined: DIRANA 1, $\sigma_{mod} = 0,2$ pS/m 1975, 266 MVA, 420/21/21 kV, Ii0, OFWF, 25°C, single phase transformer, scheduled for scrapping W_{Oil} 16 ppm at 20°C, $W_{Pap,IEH}$ 1,6 %, Original oil refilled by aged, dried oil under vacuum, cellulose dried because of vaccum	2 1,3 1,4 1,3 1 0 Mods Dira PPM RS 5 7,0 4 3,0 1,6 2,0 1 0 1 0 1 0 1 0 1 0 1 0 1 0 0 0 0 0 0 0

Table 7:On-site moisture measurement results obtained between 2003 and 2007,
sorted by manufacturing date

Transformer 7: Sampling: Dielectric diagnostic:	1971, 100 MVA, 220/25 kV, Yy0, OFAF, 5°C, scheduled for scrapping $W_{L, Paper}$ 4,8 % FD: IDA 200, 1000-0,00046 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0,38$ pS/m, TD: RVM 5462, t _C = 0,02-5000 s, PDC Analyzer 1Mod, SW 3.0 [Alff 2007]	5 4,8 4 - 3 2,2 2,5 2 1,8 2,2 2,5 1 ,8 0 1 0 KF-L Mods RVM PDC
Transformer 8: Sampling: Dielectric diagnostic:	1971, 100 MVA, 220/30 kV, Yd5, OFAF, 23°C, fault: short circuit LV winding – tank Not applied FD: IDA 200, 1000-0,001 Hz, analysed by MODS 1.5, $\sigma_{mod} = 0.1$ pS/m, TD: RVM 5462, t _C = 0,02-2000 s	5 4 3,7 2 1 0 Mods RVM
Transformer 9: Sampling: Dielectric diagnostic:	1967, 133 MVA, 230/115/48 kV, Yd5, OFAF, 41°C, three winding transformer 18 ppm at 29°C, $RS_{Oil} = 10$ % at 29°C FD: IDA 200, 1000-0,00046 Hz, analysed by MODS 1.5 and by Dirana $\sigma_{mod} =$ 2,5 pS/m, TD: Keithley 5617A electrometer	5 4 3 2,3 2,5 3,2 3,2 3,2 1 0 Mods Dira PPM RS
Transformer 10: Sampling: Dielectric diagnostic:	1963, 100 MVA, 220/110/20 kV, Yy0/d5, OFAF, 55-35°C, three winding transformer 23 ppm at 39°C FD: IDA 200, 1000-0,001 Hz, analysed by MODS 1.5, $\sigma_{mod} = 30$ pS/m, TD: RVM 5462, $t_{c} = 0,02-2000$ s	5 4 3 2 1,0 1 0 Mods RVM PPM
Transformer 11: Sampling: Dielectric diagnostic:	1954, 30 MVA, 104/23,4 kV, Yy0, ONAF, 25°C, scheduled for scrapping 17 ppm at 25°C, 11 % at 24°C, $\sigma_{meas} = 0.9$ nS/m at 22°C FD: IDA 200, 1000-0,0001 Hz, analysed by MODS 1.5 and by Dirana 1,3 pS/m, TD: Keithley 5617A electrometer	5 4,3 4,4 5,2 4 3 2,9 2 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Transformer 12: Sampling: Dielectric diagnostic:	1950, 30 MVA, 104/23,4 kV, Yy0, ONAF, 24°C, scheduled for scrapping 21 ppm at 24°C, 7 % at 24°C, $\sigma_{meas} =$ 0,9 nS/m at 22°C, $W_{Pap,IEH} = 2,6$ %, $W_{Pap,L} =$ 3,5% FD: IDA 200, 1000-0,00046 Hz, analysed by MODS 1.5 and by Dirana $\sigma_{mod} =$ 1,1 nS/m, TD: Keithley 5617A electrometer	5 4 3 2,6 2,6 2,9 2,5 2 1 0 5 5 2,6 2,9 2,5 2,5 2 1 0 5 5 7 5 7 5 7 7 7 7 7 7 7 7 7 7 7 7 7

Table 7 clearly indicates the increasing moisture content with increasing transformer age. New transformers have 0,4-0,8 % moisture in the solid insulation, whereas old transformers have 2,5-4 %. One old transformer (T10, 1963) seems to be relatively dry with 1 % moisture content.

Comparing the moisture analysis results of the commercial dielectric diagnostic methods RVM, PDC and FDS/Mods similar properties appear as indicated by the benchmark test on p. 67. The moisture analysis result by the RVM software SVRVM 2 V3.0 correlates with oil conductivity. The result obtained by PDC Evaluation Software 3.0 is somewhat higher than that of FDS Mods 1.5. The software developed within this work also delivers higher results than the commercial FDS Mods 1.5. The database used for analysis causes this difference, where different Karl Fischer titration parameters scaled the moisture content of the samples.

A very important parameter for data analysis is the insulation temperature. Since every power transformer has a thermometer, its measurement seams to be an easy task. However, as the table above shows, the value shown by the built-in thermometer usually differs from that measured in an oil sample at the same time, e.g. at transformer 9. In all cases the value directly measured in oil gave more reasonable results and thus should be preferred during on-site measurements. If several oil taps are available, the one providing the top oil should be used, since its temperature better correlates to the winding.

Using the measurement by moisture equilibrium diagrams the old approach of using the moisture by weight in ppm (e.g. [Oommen 1984]) always gives unreasonably high moisture contents (PPM). The moisture adsorption capacity of the specific transformer materials differs too much from the materials in the equilibrium diagram, usually because of aging. In contrast to that, the use of moisture saturation in oil (RS) gives a similar accuracy in moisture determination as dielectric diagnostic methods. A long term average as described at p. 59 further improves the result. The result derived from moisture saturation in oil especially reflects the cellulose that actively takes part in moisture migration, which is the winding paper and the large surfaces of the main insulation. This is an advantage over dielectric methods, which primarily reflect the main insulation.

As transformers 7 and 12 show, paper sampling does not inevitably help to evaluate moisture content. Usually the time between transformer opening and sampling is too long, the sample vessel might not be diffusion tight, the transportation to the laboratory too long and the titration parameters unknown (p. 23). Therefore in all cases titration by an external laboratory (KF-L) provided doubtful results. Trustworthy results only delivered self-made sampling, transportation and titration (KF-I).
Three measurements with interesting results will be discussed in more detail in the following sections. The three virgin transformers T1-T3 indicate difficulties of assessing very low moisture contents. For transformer 9 (1976) the tertiary winding was not in use resulting in water agglomeration. Transformer 12 (1950) has a very conductive insulation system which complicates moisture analysis by dielectric methods. Furthermore difficulties with paper sampling became obvious.

5.2 Case Studies of Selected Transformers

Assessment of Three Virgin Autotransformers

The goal of this moisture assessment was to evaluate the drying state of virgin transformers which reflects the manufacturing quality. An insulation diagnostics system IDA 200 [GE 2007] measured the dielectric properties of three virgin autotransformers having 450 MVA rated power (T1, T2, T3 in Table 7). Because of the winding setup of an autotransformer the measurements were taken between LV and tertiary winding (Figure 80).



Figure 80: Measurement setup for the insulation between LV and tertiary winding C_{HT} . The capacitances of the HV and tertiary winding to tank are guarded.

Figure 81 depicts the dissipation factor of the three transformers. Oil conductivity and insulation geometry determines the shape of the curves even down to 100 μ Hz. The dielectric properties of the solid insulation are hidden because of its low losses. On such a graph the discrimination between insulation geometry, oil conductivity and cellulose properties complicates data analysis (Figure 43). Information about the dissipation factor at lower frequencies would solve this problem. However a measurement in frequency domain down to 100 μ Hz already takes 11-12 h, measurement results for only one additional frequency point (here 41,6 μ Hz) would again double the time needed to 24 h. During a dielectric measurement the transformer is unavailable, which is very inconvenient. Here the solution will be a measurement in time domain with subsequent transformation of the result into frequency domain [Koch 2007.4].

The time need to acquire information down to $41,6 \,\mu\text{Hz}$ measured in time domain amounts approximately to 7 h, which is an essential improvement compared to a measurement solely in frequency domain.



However, the moisture analysis results of 0.6 / 0.5 / 0.4 % of T1 / T2 / T3 prove the high quality of the drying procedure of that transformer manufacture. The reason for the different moisture contents was the manufacturing process, where clamping after oven drying lasted for different times.

Analysis of the Three Winding Transformer 9 from 1967

A three winding transformer from 1967 should be moved to another substation. Depending on its actual condition three options arose: transportation without moisture treatment, drying or dedication for scrapping. This emphasises the importance of a reliable moisture evaluation.

Various moisture measurement methods were applied to estimate the moisture content on-site. The transformer was manufactured in 1967, had a rated power of 133 MVA, a transformation ratio of 230 / 115 / 48 kV and OFAF cooling. Two meters measured the dielectric properties of the insulation as polarization and depolarization currents (Keithley electrometer 6517A) and complex capacitance (IDA 200) between HV winding and MV winding, MV winding and LV winding and LV winding and tank. Figure 82 illustrates the measurement setup for the main insulation between HV and LV winding. To avoid currents via bushings and LV winding the transformer tank and the LV winding are connected to ground and guard. Figure 83 depicts the measurement result as complex capacitance for the three insulation systems HV-MV, MV-LV and LV-tank.



Figure 82: Measurement setup for the main insulation C_{HL} . The LV winding is here connected to ground/guard.

Figure 83: Complex capacitance between HVwinding and MV-winding C_{HL} , MV-winding and LV-winding C_{LT} and LVwinding and tank C_T

The new program and Mods 1.5 [GE 2007] analyzed the measurements and came to the results in Figure 84. A capacitive probe measured the relative moisture saturation in oil on-site and the moisture in cellulose was derived via moisture isotherms (p. 57). Beside this, coulometric Karl Fischer titration determined the moisture by weight (ppm) in an oil sample and the equilibrium diagram published by [Oommen 1984] served to show moisture in cellulose. Ideally all the methods should come to a similar moisture value in cellulose.

Indeed the analysis software Mods 1.5 and the new software showed similar results, minor discrepancies arise from the different data pools. The higher moisture content in the LV insulation agrees well with the service conditions of the transformer: the LV winding was not in use. Cellulose at lower temperatures stores the water in a transformer. Thus the dielectric methods allow for an elementary localisation of wet areas in the insulation. Contrary to this the moisture content in cellulose as derived from oil samples gives an average value. The result obtained from a moisture sorption isotherm (p. 57) agrees well

with the dielectric analysis. However it must be noticed, that the measurement between LV winding and tank proceeded without guarding and thus, because of currents through the bushings, the moisture content was probably overestimated.



Figure 84: Moisture content in the solid insulation as obtained from Mods 1.5, the new analysis software (Dirana) and via equilibrium diagrams from the relative saturation of oil (RS) and from moisture by weight in oil (PPM)

For the equilibrium approaches the conventional method of deriving moisture in cellulose from moisture by weight in oil (ppm) gives a unreasonable high result. Aging of oil and paper makes the application of equilibrium diagrams from literature sources impossible in most cases.

The example of this transformer shows that a moisture distribution can be detected by dielectric measurements applied to different winding insulations. The equilibrium approach used here will only show an integral result.

Finally the operators of this transformer decided for on-site drying by oil circulation for several months. After this the transformer will be moved to another substation.

Analysis of the Heavily Aged Transformer 12

Transformer 12 was destined for scrap and such a case gives a good opportunity for comparing different approaches in assessing moisture content. Paper samples were taken out after measuring the dielectric properties (polarization and depolarization currents with Keithley electrometer 6517A, complex capacitance with IDA 200) and oil sampling (moisture content in ppm, moisture saturation in %). The insulation temperature was 24°C.

Figure 85 displays the polarisation and depolarisation currents through the main insulation as a superposition of conductivity and polarisation phenomena (p. 73). The polarisation current approaches its final value quite early; at times above 300 s almost no further decrease occurs. The polarization processes decayed and the character of the current becomes solely conductive. Since the final current value can be estimated at 300 s, the measurement might have been stopped at that time. A comparison to the common shape of polarisation currents (Figure 42) illustrates the unusually high conductivity of this transformer.





Figure 86 displays the dissipation factor as measured between HV- and LVwinding. The losses in the insulation are very high with e.g. 0,13 at 50 Hz. Also the comparison to the usual shape of the curve in Figure 43 makes the conductive nature of this insulation clearer. In the graph below oil conductivity influences the dielectric properties only down to 0,1 Hz, below this frequency the properties of cellulose dominate. Because of the highly conductive nature of this insulation the measurements might have been stopped at 0,01 Hz, where sufficient information for analysis were gathered.



Figure 86: Dissipation factor as measured between HV- and LV-winding at 140 V_{RMS}

The oil conductivity of this transformer was measured in an oil sample and modelled by the dielectric diagnostic methods, giving $\sigma_{meas} = 0.9$ nS/m at 22°C and $\sigma_{mod} = 1.3$ nS/m at 24°C. Both results agree very well, as observed for all comparisons of measured to modelled oil conductivities. The measured oil conductivity and likewise the neutralisation value of 0.49 g KOH / kg oil were unusually high as the oil conductivity at room temperature normally amounts 0.05-20 pS/m. This high value indicates a strong influence of conductive aging products and therefore a progressive aging state.



Figure 87: Moisture content in the solid insulation as obtained from Karl Fischer titration by an external laboratory (KF-L) and own measurements (KF-I), dielectric analysis: Mods 1.5, the new analysis software (Dira), PDC and via equilibrium diagrams from moisture by weight in oil (PPM) and also from the relative saturation of oil (RS)

Figure 87 compares the moisture results of the different measurement and analysis approaches. Karl Fischer titration was done by an external laboratory and by the laboratory of the IEH, Universitaet Stuttgart. The external laboratory measured nine samples taken from three different positions. The results for the three positions were 4,3-6,0; 4,2-6,4 and 3,0-3,8 % moisture by weight. The samples from the first and the second position were regarded as unreasonably wet for the moisture contents indicated equilibrium with the surrounding air. The sampling conditions were unknown. Thus Figure 87 displays the samples from the third position only but still the results are questionable. Titration in the laboratory of the IEH came to 2,6 % moisture by weight, which is distinctly low.

The analysis results of the dielectric methods also differ. Two algorithms, the commercial Mods 1.5 and self-made PDC evaluation, had no compensation for conductive aging products and came to 3,8 and 4 % moisture by weight. Within this work the developed software Dirana compensates for conductive aging products (p. 104) and indicates 2,9 % moisture relative to weight. More tests are needed to fully verify the new software.

In an oil sample the moisture saturation was measured directly on-site and also the moisture content in ppm by direct titration at the IEH laboratory. Via a moisture sorption isotherm the relative saturation gave a reading of 2,5 % moisture content in cellulose, which agrees well with the paper sample measured at the IEH (KF-I) and the new analysis software Dirana (Dira). A traditional equilibrium chart [Oommen 1984] gives a too high moisture content of 6,0 %, showing again that diagrams not adapted to aging state and moisture adsorption capacity won't help to evaluate moisture in power transformers.

To conclude, the findings for this very aged transformer 12 show, that a compensation for aging products is a necessity both for the measurement based on moisture equilibrium and that based on dielectric properties.

5.3 Influence of Moisture Migration

Moisture Non-Equilibrium

Dielectric response measurements often take place immediately after switching off a transformer. Thus the temperature decreases during the measurement, resulting in a moisture migration from oil to cellulose. During the CIGRÈ main session 2006 in Paris this topic caused the special reporter to ask "Is it possible to set the measuring condition of tan δ to the same condition as the transformer while in operation?" [Gulski 2006]. This section tries to quantify the deviations which must be expected under non-equilibrium conditions.

A Practical Example

Figure 88 illustrates the temperature decrease during the measurement at transformer 10 in Table 7 having OFAF cooling. The transformer was switched off at 08:30 am, the FDS measurement run from 10:30 am to 02:00 pm, followed by RVM to 06:00 pm and by PDC to 02:30 am of the following day. Until 09:00 am of the following day the temperature displayed at the build-in thermometer decreased from 59 to 35°C. All cooling systems were switched off during this time.



Because of temperature decrease and moisture migration two influences on the dielectric can be expected: an elevated oil conductivity and water attraction at the surfaces of the cellulose insulation.

Influence of Oil Conductivity during Temperature Decrease

At decreasing temperatures the moisture solubility in oil *decreases*, whereas that of cellulose material *increases*. Thus a temperature decrease in an oil-paper-insulation causes moisture migration from oil to cellulose. While the equilibration process is running, the moisture in oil is higher than that of equilibrium. According to Figure 53 oil conductivity depends on moisture in oil.

This follows in oil conductivity which is too high at non-equilibrium. On the other hand, due to the decreasing temperature the oil conductivity will *decrease*.

Figure 89 helps to quantify the interaction of the various influences in the real case of transformer 10 of Table 7. To emphasize the influence of moisture, a moisture content in paper of 2 % is assumed. Under equilibrium conditions this results in a moisture saturation of 6 % in oil, derived from the isotherms in Figure 20. The moisture content (ppm) in oil will then come to 14 ppm at 60°C (Table 3, Nynas Nytro 3000). Now the most critical case will be supposed: no moisture migration occurs. Thus the moisture content will remain constant, but the moisture saturation will increase to 13 % at the end of the measurement sequence (38°C oil temperature). Because of the increased moisture saturation the oil conductivity will increase from 30 to 36 pS/m (Figure 53). At the same time because of the temperature decrease the conductivity will decrease to 9 pS/m, calculated using equation (35).



Conclusively the influence of moisture nonequilibrium on oil conductivity will be overlaid by the temperature dependence of oil conductivity. Additionally, well programmed analysis algorithms are able to compensate for the influence of oil conductivity (p. 67).

Water Attraction at the Surfaces of the Cellulose Insulation

The second effect of moisture migration on dielectric measurements consists in the agglomeration of water molecules at the surfaces of paper and pressboard. To judge the relevance of this phenomenon a practical example may help. During the comparative test on dielectric diagnostic methods as depicted at p. 63 a "pancake model" was used, featuring insulation structures similar to real power transformers. With a weight of 1,3 t and 270 l of oil its thermal time constant is approximately 15 h. The dielectric properties of the insulation were measured during cooling down, immediately after reaching room temperature and five months later.

Figure 90 depicts the polarisation current flowing through the model at decreasing temperature for a time span of 24 h. The current follows the temperature progression very closely. After reaching a stable temperature no change of the current appears. If moisture migration influenced the dielectric properties, the current should alter for a longer period.



In a second experiment the dielectric properties of connection DG-CH (ratio of pressboard to oil 25/75 %) were measured immediately after cooling down from 80 to 21°C and five months later, when equilibrium should have been reached. Supposing moisture non-equilibrium influences the dielectric properties, then a difference should be evident between the first and the second measurement.

Figure 91 depicts the capacitance of the model as real and loss part. Indeed the figure indicates a minor increase at the loss part for frequencies below 21 mHz. However, for moisture evaluation with an analysis software this increase has no influence. The persistent difference between measured and modelled curve hides such a minute variation.



Conclusion and Quantification

Reasonable physical influences caused by moisture non-equilibrium are oil conductivity which is too high and moisture agglomeration at the surfaces of the cellulose insulation. After closer consideration both are of slight effect, since they are hidden by the decreasing temperature or have a negligible impact. An appropriate appraisal for this deviation seems to be below 10 % compared to the moisture analysis result at equilibrium.

6 Summary, Conclusions and Suggestions

This thesis aims to provide reliable methods for moisture assessment of oilpaper-insulated power transformers. For a suitable basis it investigates direct moisture measurements by Karl Fischer titration and capacitive probes. Then indirect approaches by means of moisture equilibrium are introduced. This is followed by dielectric measurements, where moisture in the solid insulation is derived from dielectric properties. Moisture determination of real power transformers complete the work.

6.1 Summary and Achievements

Condition Based Maintenance Requires Reliable Diagnostic Tools

Condition based maintenance strategies require reliable information about the actual state of electrical equipment. Power transformers represent the most expensive links in the chain connecting generation to utilization. Due to cost pressure of a liberalized energy market the utilities shift maintenance from time based to condition based approaches. This development requires reliable diagnostic tools.

Water in oil-paper-insulations causes three detrimental effects: decrease in dielectric withstand strength, accelerated cellulose aging and the emission of bubbles at high temperatures. Therefore knowledge about the actual moisture concentration is required in order to decide on further action such as, for example, on-site drying. However, the commercially available measurement techniques often provide conflicting results.

Based on the uncertainties of conventional techniques currently used, this work has the objective of enhancing knowledge about water sorption and equilibrium, explaining and eliminating errors of conventional moisture measurement methods, describing and measuring moisture in power transformers by means of the relative saturation, improving interpretation schemes for dielectric diagnostic methods and finally giving correlations of new approaches to traditional methods.

Direct Measurements by Karl Fischer Titration and Capacitive Probes

The titration according to Karl Fischer represents the state of the art method for moisture measurements directly in oil and paper. However a round robin test revealed poor comparability between different laboratories especially at oil samples (p. 25).

To improve this, the findings of this work propose for titrations on cellulose samples a heating temperature of 160° C for paper and 180° C for pressboard. This not only releases more water but ensures a higher reproducibility and shorter titration time as well. To part cellulose from oil, heptane performs much better than the often used methanol (p. 27). For titration of aged oils the direct injection of the oil sample into the titration cell gave better results than external heating. For direct titration the influence of aging by-products was found to be below 10 % (p. 28). The appendix at p. 127 describes a recommended procedure for reliable Karl Fischer titration.

A much easier moisture measurement method gives the determination of relative humidity using capacitive probes. Saturated salt solutions easily determine their high accuracy. The probes performance proved a comparison to chilled mirror dewpoint hygrometers (p. 31).

Special attention to the frequent discussion comparing Karl Fischer titration to capacitive probes was given on p. 32. It follows, that due to the different measurement principles (chemical reaction vs. moisture deposition) a comparison is impossible without calibrating at least one of the principles to the other.

Measurement through Moisture Equilibrium

On-site moisture determination using Karl Fischer titration for oil samples with subsequent application of an equilibrium diagram is still frequently used by transformer operators. However, this approach is subject to severely disturbing influences as for example sampling and titration procedure, equilibrium conditions and particularly aging (p. 36). For condition assessment as a basis of further maintenance this approach does not deliver the desired accuracy.

Although the conventional approach gives very disappointing results, the achievements of the work presented here led to moisture equilibrium evolving into the favoured one for on-line moisture determination in transformers.

To reach this, at first the physical background of moisture adsorption in cellulose and moisture solubility in oil was described (pp. 39, 45). Then moisture isotherms for paper and pressboard were recorded, describing the ratio of water content in the material to the exerted water vapour pressure (water saturation). Cellulose materials have a typical s-shaped isotherm, indicating that water molecules are strongly bound for low concentrations and become exponentially more available for increasing moisture content (> 1,5 %). The moisture adsorption capacity of insulation paper and pressboard are similar, but not identical (p. 42). Aging decreases the moisture adsorption capacity, probably because of hornification.

Moisture isotherms for mineral and vegetable oil were also measured (p. 46). Oils in contrast to cellulose feature a linear relation between moisture content and moisture saturation. For mineral oils, aging increases moisture solubility, which is opposite to the behaviour of cellulose (p. 47). A correlation between moisture solubility and acidity was observed. Vegetable insulation oils can take up 20-40 times more water than mineral oil.

Moisture migration and equilibrium are determined by differences of relative humidity. The theoretical background for moisture measurements through equilibrium provided considerations about thermodynamic equilibrium (p. 49). The relations between water potential, water vapour pressure and relative humidity were given. An experiment verified practically the theory on moisture equilibrium (p. 51).

Based on this knowledge isotherms for cellulose and oil were combined in order to achieve equilibrium diagrams relating the water mass in cellulose to that in oil (p. 53). Since these equilibrium diagrams are adapted to the moisture adsorption capacity of the involved materials, they feature a higher reliability than the ones from traditional literature sources. Nevertheless, they have the essential drawback in that they have to be adapted to every material and its aging state.

An essential improvement here gives the use of moisture saturation in oil instead of moisture content in ppm (p.57). In this way, the moisture solubility of the specific oil becomes negligible, no matter whether it is an aged mineral or a vegetable oil. Moisture isotherms for wooden transformer materials can now be used to derive moisture by weight in the solid insulation from moisture saturation in oil. One drawback still remains: the moisture adsorption capacity of cellulose varies with different materials and decreases with aging.

The solution of this problem leads to the key recommendation of this thesis: to use water saturation in oil *and in paper* to determine water in power transformers. All destructive effects of water correlate better with water saturation than with the traditionally used moisture relative to weight. Also for drying purposes water saturation serves better since it reflects the available water molecules. Additionally, capacitive probes on-line determine water saturation in oil and, with a long time average, in cellulose as well (p. 59). Also dielectric diagnostic methods can determine water saturation (p. 105).

Moisture Analysis by Dielectric Diagnostic Methods

Dielectric response methods were developed with the promise of more certain results than the hitherto used equilibrium method based on moisture content in oil. However the first approach, the recovery voltage method RVM, soon appeared to depend on oil conductivity and not on moisture content. The newer approaches which are polarisation / depolarisation currents PDC and frequency domain spectroscopy FDS provided more reliable results. However, a

benchmark test also revealed weaknesses in compensating for insulation temperature and oil conductivity (p. 67). Furthermore the conventional approaches not regard the influence of conductive aging by-products on the dielectric properties.

The theory on conductivity and polarisation of materials points out, that measurements on dielectrics always imply a superposition of the phenomena interfacial polarisation, conductivity, moisture, conductive aging by-products, and temperature (p. 73). Since dielectric measurements aim to assess moisture, the quality of analysis consists in a separation of the different effects.

The dielectric properties of oil and of pressboard were measured in time and frequency domain in order to get a reliable data pool for moisture assessment. Mineral oils show a time and voltage dependent conductivity, influenced by charge clouds in the measurement cell. The conductivity of new oils is around 0,05 pS/m, for very aged it increases to 1300 pS/m, showing a slight correlation to acidity (p. 83) and to moisture saturation. The temperature dependence of oil conductivity can be described by activation energy, which ranged from 0,32 to 0,54 eV.

In frequency domain the dielectric properties of pressboard models were particularly specially measured. The influences of moisture, aging and insulation temperature were investigated as well as that of pressboard type and oil impregnation (p. 91). Moisture increases losses and polarizability and aging behaves very similar to this. Furthermore it was found that high density pressboard materials (1,1-1,3 g/cm³) have a higher polarizability ($\varepsilon' = 4,6$) compared to the low density materials (0,9 g/cm³) having $\varepsilon' = 3,0$. The losses, the imaginary part ε'' of permittivity, are higher for the high density materials as well. For analysis of a real power transformer this results in an uncertainty of the obtained moisture content (p. 97).

A new moisture analysis algorithm was developed (p. 101). Measurements on pressboard and oil formed a database for on-site moisture assessment in power transformers. Moisture analysis bases on a comparison of modelled dielectric properties to measured dielectric properties of real transformers. The analysis algorithm features weighting of low frequency data, an extended XY-model and compensation for conductive aging by-products.

On-Site Measurements at Power Transformers

Measurements on power transformers were made verify the findings of this work and compare conventional to new approaches.

For the equilibrium approach, the traditional application of equilibrium diagrams in all cases led to excessively high moisture contents in paper. Contrary to this equilibrium diagrams in the shape of moisture isotherms always delivered credible results.

The dielectric diagnostic methods PDC and FDS performed well in most cases. Differences in the analysed moisture content are mainly caused by different data bases scaled with different Karl Fischer titration techniques. One drawback of the conventionally available dielectric diagnostic methods is their incapacity to compensate for conductive aging by-products leading to an apparent excessive moisture content for aged transformers (p. 114). The software developed within this work successfully compensates this influence, however more test results need to be gathered.

Practical examples depicted the influence of moisture migration on dielectric response methods (p. 117). It was found, that the conceivable disturbances by oil conductivity and water attraction at surfaces are hidden by the temperature dependence of dielectric properties.

In conclusion, the analysis of dielectric properties allows for a reliable moisture determination in power transformers provided that temperature, insulation geometry, oil conductivity and aging by-products are accordingly compensated. Assessing moisture via moisture equilibrium is very easy feasible, accurate and applicable on-line. Using moisture saturation it features a measure that directly relates to drying and to the destructive effects of moisture in oil-paper-insulations. Maintenance actions should base on consistent indications by both, moisture equilibrium and dielectric diagnostic methods.

6.2 Suggestions for Further Work

Since Karl Fischer titration represents the basis for moisture content measurements, improvements on this technique should have the highest priority when suggesting further research. The current state of the art consists of a poor comparability between different laboratories and for oil samples even the reproducibility within one laboratory has room for improvement. Although this thesis gives some attempts for more reliable titrations, much more comprehensive work needs to be done in order to improve Karl Fischer titration or to replace it by another method for determining water mass.

Capacitive probes feature a much higher accuracy but their long term stability and interferences by aging by-products still need to be examined.

At moisture isotherms of cellulose and oil the effect of aging should be investigated more closely. Differences between artificial and real life aging procedures became obvious. More practical experiences are required for measurements of relative saturation. Beyond this the influence of relative saturation on aging rate needs further investigation. Dielectric diagnostic methods may in future be based on a data pool measured at non-impregnated paper. In this way the properties of the impregnating oil become negligible, however in this case a new scheme of interpretation is required. The approach to compensate for conductive aging by-products as proposed within this work needs to be verified by more laboratory and field measurements. An interesting field of research is the connection between dielectric properties of cellulose, oil conductivity and oil breakdown voltage with special attention to low molecular acids.

7 Appendix

7.1 Zusammenfassung

Die vorliegende Dissertation beabsichtigt, zuverlässige Messmethoden für Feuchtigkeit in Öl-Papier-isolierten Transformatoren bereitzustellen. Dazu werden im ersten Schritt Methoden zur *direkten* Wasserbestimmung in Öl und Papier vorgestellt und deren Zuverlässigkeit sichergestellt. Danach werden als neue, *indirekte* Messmethoden die Feuchtebestimmung in Öl und Zellulose (Papier, Pressboard) mittels Gleichgewichtsverfahren und die dielektrische Diagnostik vorgestellt und schrittweise verbessert. Schließlich zeigen Messungen an Leistungstransformatoren die praktische Anwendung der neuen Verfahren im Vergleich zu traditionellen Methoden.

Zustandsbasierte Instandhaltung verlangt zuverlässige Diagnostik

Zustandsbasierte Instandhaltungsstrategien versprechen gegenüber der traditionellen zeitabhängigen Instandhaltung nur dann Vorteile, wenn der wirkliche Zustand des Betriebsmittels sicher ermittelt wurde. stellen das teuerste Leistungstransformatoren Glied der in Energieübertragungskette vom Generator zum Verbraucher dar. Der Kostendruck des liberalisierten Energiemarktes lässt die Energieversorgungsunternehmen auf statt kostensparende zustandsabhängige Verfahren auf traditionelle zeitabhängige Instandhaltung setzen. Dieser Ansatz verlangt zuverlässige Diagnoseverfahren.

Feuchtigkeit in Öl-Papier-Isolierungen ist ein wesentliches Zustandsmerkmal. der Feststoffisolierung beträgt 0,4-1 % Der Wassergehalt für neue Transformatoren und steigt auf 3-4 % am Ende der Lebensdauer von etwa 30-50 Jahren. Das entspricht einer Wassersättigung von 0,5-2 % im Neuzustand und bis zu 25 % für feuchte Transformatoren. Feuchtigkeit schädigt durch drei Wirkungen: Die Durchschlagsspannung von Öl und Zellulose wird verringert, die Alterung der Zellulose beschleunigt und die Emission von Gasblasen bei hohen Temperaturen verursacht. So ist die Kenntnis der Feuchtigkeit im Transformator von hoher Bedeutung für einen sicheren Betrieb und zustandserhaltende Maßnahmen. Trotz dieser Zusammenhänge führen kommerziell erhältliche Messverfahren für Feuchtigkeit häufig zu widersprüchlichen Ergebnissen, selbst eine Klassifikation in trocken. mittelmäßig und feucht ist nur mit beschränkter Sicherheit möglich.

Dieser Stand der Technik motiviert zur vorliegenden Dissertation. Ihre Ziele sind:

- das Wissen über Sorption, Diffusion und Gleichgewicht von Feuchtigkeit zu vertiefen und dabei Alterung zu berücksichtigen,
- eine neuen Ansatz zur Beschreibung und Messung von Feuchtigkeit in Transformatoren einzuführen: die Wassersättigung für Öl und Zellulose (Papier, Pressboard),
- Fehler konventioneller Methoden zu erklären und zu eliminieren, z.B. bei der Titration nach Karl Fischer und bei Gleichgewichtsmethoden,
- die Interpretation dielektrischer Methoden zu verbessern, z.B. durch Berücksichtigung der transienten Eigenschaften der Ölleitfähigkeit und der Wirkung leitfähiger Alterungsprodukte,
- einen zuverlässigen Datensatz zur Analyse dielektrischer Messungen bereitzustellen,
- auch Bedienern ohne Expertenwissen reproduzierbare Ergebnisse über einen langen Zeitraum zu ermöglichen,
- die Vergleichbarkeit zwischen neuen und traditionellen Ansätze zu sicherzustellen z.B. durch Gegenüberstellung bei Vorort- und Werkstattmessungen und schließlich
- Feuchtigkeit so genau beurteilen zu können, dass ein zuverlässiges Kriterium für weitere Instandhaltungsmaßnahmen bereitsteht. Eine Genauigkeit von ± 15 % des Analyseergebnisses genügt z.B. für eine Entscheidung über eine Trocknung.

Direkte Messung durch Karl Fischer Titration und Kapazitive Sensoren

Die Titration nach Karl Fischer repräsentiert den Stand der Technik für *direkte* Feuchtemessungen in den Materialien Öl und Zellulose. Jedoch bewiesen mehrere Blindtests eine unbefriedigende Vergleichbarkeit zwischen verschiedenen Laboratorien, besonders für Ölproben (S. 25).

Die Ergebnisse dieser Arbeit lassen nach eingehenden Untersuchungen eine Ausheiztemperatur von 160°C für Papier und 180°C für Pressboard als am geeignetsten erscheinen. Diese setzt nicht nur mehr Wasser frei sondern erhöht auch die Reproduzierbarkeit und verkürzt die Titrationszeit. Zur Trennung von Öl und Zellulose eignet sich Heptan wesentlich besser als das häufig benutzte Methanol (S. 27). Bei der Titration von gealterten Ölen erwies sich die direkte Probeninjektion als zuverlässiger gegenüber dem Ausheizen des Öls. Bei der direkten Injektion kann der Störeinfluss durch Alterungsprodukte im Öl mit unter 10 % angegeben werden (S. 28). Der Anhang auf S. 127 beschreibt eine empfohlene Vorgehensweise für Karl Fischer Titrationen. Wesentlich einfacher gestaltet sich die Messung der relativen Feuchtigkeit mittels kapazitiver Sensoren. Hier können gesättigte Salzlösungen einfach die Genauigkeit überprüfen. Außerdem bewies ein Vergleich zu Spiegelhygrometern die Genauigkeit und guten dynamischen Eigenschaften kapazitiver Sensoren.

Den häufig diskutierten Diskrepanzen zwischen Titration nach Karl Fischer und kapazitiven Sensoren galt besondere Aufmerksamkeit (S. 32). Hier machen die unterschiedlichen Messprinzipien (chemische Reaktion gegenüber Wasseradsorption) einen direkten Vergleich unmöglich. Dessen Voraussetzung ist es, ein Prinzip auf das andere zu kalibrieren.

Messung durch Feuchtegleichgewicht

Bei der Vorort-Feuchtebestimmung stellt die Karl Fischer Titration von Ölproben mit nachfolgender Anwendung eines Gleichgewichtsdiagramms immer noch die am häufigsten genutzte Methode dar. Dennoch beschränken verschiedene Einflüsse wie Probenentnahme, Titration, Gleichgewichtsbedingungen und besonders Alterung deren Genauigkeit (S. 36). Völlig unzureichende Ergebnisse liefert sie zur Zustandsbestimmung als Basis nachfolgender Instandhaltungsmaßnahmen. des enttäuschenden Trotz herkömmlichen Gleichgewichtsverfahrens machen die Ergebnisse dieser Arbeit das Prinzip Feuchtegleichgewicht zur favorisierten Methodik zur Feuchtebestimmung in Transformatoren.

Für dieses Ergebnis wurden zunächst der physikalische Hintergrund von Wasseradsorption und –löslichkeit in Zellulose beschrieben (S. 39, 45), danach Sorptionsisotherme für Papier und Pressboard experimentell ermittelt. Sie beschreiben den Zusammenhang zwischen Wassergehalt und Wassersättigung (Wasserdampfdruck). Auf Zellstoff basierende Materialien besitzen s-förmige Isotherme, welche eine starke Wasserbindung bei niedrigen Wassergehalten und eine exponentiell höhere Verfügbarkeit bei höheren Gehalten anzeigen(> 1,5 %). Die Wasseraufnahmefähigkeit von Pressboard und Papier stellt sich ähnlich dar (S. 42). Alterung verringert die Wasseraufnahmefähigkeit, wahrscheinlich wegen des Effekts der Verhornung.

Sorptionsisotherme wurden ebenfalls für Mineral- und Pflanzenöle gemessen (S. 46). Im Gegensatz zu Zellulose zeigen diese eine *lineare* Verknüpfung zwischen Wassergehalt und Wassersättigung. Alterung bewirkt bei Mineralölen eine erhöhte Wasserlöslichkeit (S. 47), was ebenfalls gegensätzlich zur Zellulose ist. Weiter wurde ein gewisser Zusammenhang zwischen Wasserlöslichkeit und Säuregehalt des Öls beobachtet. Pflanzliche Isolieröle nehmen eine etwa 20-40-fache Wassermenge gegenüber Mineralöl auf.

Wasserdiffusion und –gleichgewicht werden durch Unterschiede in der relativen Feuchtigkeit bestimmt. Feuchtemessungen mittels Gleichgewicht beruhen auf dem thermodynamischen Gleichgewicht. Die Beziehungen zwischen Wasserpotential, Wasserdampfdruck und relativer Feuchtigkeit wurden beschrieben und ihre praktische Anwendbarkeit experimentell überprüft (S. 51).

Darauf basierend konnten Sorptionsisotherme von Zellstoff und Öl verknüpft werden, was zu Gleichgewichtsdiagrammen führt, welche die Wassermasse in Öl auf die Wassermasse in Zellstoff beziehen (S. 53). Da diese Diagramme für die wirklich verwendeten Materialien gelten, bieten sie eine höhere Genauigkeit als Diagramme herkömmlicher Literaturquellen. Dennoch tragen sie den wesentlichen Nachteil, dass sie für jedes Öl-Papier-System angepasst werden müssen.

Zu einer deutlichen Verbesserung führt die Verwendung der Wassersättigung des Öls statt des Wassergehalts in ppm (S. 57). Die Wasserlöslichkeit des Isolieröls muss dann weder für ein Mineral- noch für ein Pflanzenöl berücksichtigt werden. Sorptionsisotherme von Zellstoffmaterialien bestimmen nun die Gewichtsfeuchte in der festen Isolierung aus der Wassersättigung des umgebenden Öls. Dennoch bleibt auch hier ein Nachteil, die Sorptionskapazität von Zellstoff ist materialspezifisch und verringert sich mit der Alterung.

Die Lösung dieses Problems führt zur Schlüsselthese dieser Dissertation: Wassersättigung in Öl und *ebenfalls im Papier* zur Bewertung der Feuchtigkeit von Transformatoren zu benutzen. Alle schädigenden Effekte von Wasser korrelieren besser mit der Wassersättigung als mit dem traditionell benutzten Wassergehalt (Gewichtsfeuchte). Auch für Trocknungsprozesse ist die Wassersättigung besser geeignet, da sie das verfügbare Wasser charakterisiert. Darüberhinaus messen kapazitive Sensoren einfach und genau auch während des Transformatorbetriebs (S. 59). Die Wassersättigung kann ebenfalls durch dielektrische Diagnoseverfahren bestimmt werden (S. 105).

Feuchtebestimmung durch Dielektrische Diagnostik

Zur Zeit ihrer Einführung versprachen dielektrische Diagnosemethoden eine höhere Genauigkeit als die damals übliche Gleichgewichtsmethode basierend auf der Gewichtsfeuchte in Öl (ppm). Dennoch wurde bald offenbar, das die zuerst kommerzialisierte Wiederkehrspannungsmethode (Recovery Voltage Method RVM) zu stark von der Ölleitfähigkeit beeinträchtigt wurde. Als viel zuverlässiger erwiesen sich die neueren Ansätze der Polarisationsstrommessung (Polarisation / Depolarisation Currents PDC) und der frequenzabhängigen Verlustfaktormessung (Frequency Domain Spectroscopy FDS). Dennoch offenbarte ein Vergleichstest auch hier Schwächen bei der Kompensation von Ölleitfähigkeit und Isolationstemperatur (S. 67). Zudem kann keine der leitfähiger herkömmlichen Methoden den Einfluss Alterungsprodukte kompensieren (z.B. Säuren), die Feuchtigkeit stark gealterter Transformatoren wird dann überschätzt.

Messungen an einem Dielektrikum implizieren immer eine Überlagerung der Grenzflächenpolarisation und Effekte Leitfähigkeit, wie theoretische Betrachtungen zeigen (S. 73). Diese wiederum werden bestimmt durch leitfähige Alterungsprodukte Feuchtigkeit. und Temperatur. Da Feuchtebestimmung das Ziel der Messung ist, besteht die Qualität einer Feuchtigkeitsanalyse in der Unterscheidung dieser Effekte.

Die Feuchtigkeitsanalyse beruht auf einem Vergleich zwischen einer modellierten dielektrischen Antwort aus der Datenbasis und der dielektrischen Antwort des Leistungstransformators. Um eine zuverlässige Datenbasis zu erhalten, wurden die dielektrischen Eigenschaften von Öl und Pressboard im Zeit- und Frequenzbereich gemessen. Mineralöle zeigen eine zeit- und frequenzabhängige Leitfähigkeit, beeinflusst von Ladungsträgerwolken in der Messzelle. Die Leitfähigkeit neuer Öle liegt bei 0,05 pS/m, steigt durch Alterung auf mehr als 1000 pS/m und korreliert leicht mit Wassersättigung und Säurezahl (S. 83). Die Temperaturabhängigkeit der Ölleitfähigkeit kann mittels der Aktivierungsenergie beschrieben werden, welche hier bei 0,32 bis 0,54 eV lag.

Dielektrische Messungen an Pressboard-Proben im Frequenzbereich wurden besonders umfangreich durchgeführt um den Einfluss von Feuchtigkeit, Alterung, Temperatur, Pressboardtyp und Imprägnierung zu bestimmen (S. 91). Feuchtigkeit erhöht die Verluste und Polarisierbarkeit, davon nicht unterscheidbar verhalten sich leitfähige Alterungsprodukte. Pressboard mit höherer Dichte (1,1-1,3 g/cm³) weist eine höhere Permittivität auf ($\varepsilon' = 4,6$) als Material niedrigerer Dichte (0,9 g/cm³) mit $\varepsilon' = 3,0$. Bei der Analyse von Leistungstransformatoren können diese Eigenschaften die Genauigkeit beeinträchtigen (S. 97).

Zur Auswertung von Messungen an Leistungstransformatoren wurde ein neuer Algorithmus entwickelt (S. 101). Die Messungen an Öl und Pressboard bekannter Feuchte bilden eine Datenbasis. Der Analysealgorithmus gewichtet die Niederfrequenzdaten, benutzt ein erweitertes Geometriemodell, kompensiert leitfähige Alterungsprodukte und bietet neben dem Wassergehalt auch die Wassersättigung als Ergebnis.

Vorort-Messungen an Leistungstransformatoren

Durch Vorort-Messungen sollen die neu entwickelten Verfahren überprüft und mit herkömmlichen verglichen werden. Das traditionelle Gleichgewichtsverfahren basierend auf dem Wassergehalt im Öl in ppm lieferte in jedem Fall zu hohe Wassergehalte in der festen Isolierung. Im Gegensatz dazu erzielten Gleichgewichtsdiagramme in der Form von Sorptionsisothermen immer glaubwürdige Ergebnisse.

Die dielektrischen Methoden PDC und FDS erwiesen sich in den meisten Fällen als geeignet. Unterschiede im ermittelten Wassergehalt können mit den unterschiedlichen Vergleichsdatensätzen, skaliert mit unterschiedlichen Karl Fischer Titrationsverfahren, begründet werden. Als Nachteil der konventionellen Methoden zeigten sich die lange Messzeit bei der Frequenzbereichsmessung FDS und die mangelnde Möglichkeit, leitfähige Alterungsprodukte zu kompensieren. Dies führte im Fall eines stark gealterten Transformators zu überschätztem Wassergehalt (S. 114). Die in dieser Arbeit vorgestellte Software konnte den Einfluss leitfähiger Alterungsprodukte kompensieren, jedoch sind hier weitere praktische Messungen nötig.

Der Einfluss von Feuchtemigration auf dielektrische Diagnosemethoden wurde anhand von Berechnungen und praktischer Messungen quantifiziert. Die denkbaren Störungen durch ein Feuchteungleichgewicht während sinkender Temperatur werden durch die Temperaturabhängigkeit der dielektrischen Eigenschaften überdeckt (S. 117).

Zusammenfassend bewerten dielektrische Diagnosemethoden zuverlässig die Feuchtigkeit in Leistungstransformatoren, wozu jedoch überlagernde Einflüsse durch Ölleitfähigkeit, Temperatur und Alterung kompensiert werden müssen. Feuchtebestimmungen über das thermodynamische Gleichgewicht sind einfach ausführbar, genau und im Betrieb durchführbar. Mit der Wassersättigung bietet sich eine Messgröße an, die direkt mit der Trocknung und den zerstörerischen Effekten von Feuchtigkeit in Öl-Papier-Isolierungen verknüpft ist. Instandhaltungsmaßnahmen sollten auf übereinstimmender Indikation durch beide Verfahren beruhen, dem Feuchtegleichgewicht und der dielektrischen Diagnostik.

7.2 Recommendations for Karl Fischer Titration

On-Site Sampling

The sampling procedure determines the accuracy of the moisture analysis so it should be performed with care.

Oil should be sampled from different levels in the transformer; if possible top, middle and bottom. Samples should not be taken in high air humidity (rain, fog). When taking samples, a few litres of oil should be drained out so preventing sludge and standing oil to be sampled. The bottle should be dry and technically clean that moisture inside the bottle cannot contaminate the oil. For the same reason the bottle should be completely filled, leaving no space for air at the top. Since transformer oil is considered to be harmful to the environment, precautions must be taken to avoid contamination of the soil around the transformer.

Paper should be sampled as soon as possible after opening the transformer. The outer layers of the conductor insulation may be moistened by air, thus it should

be removed. Remove the edge of paper and pressboard samples also for the same reason. The transformer insulation oil is useful for storing paper and pressboard samples during transportation to the laboratory. Besides the cellulose samples keep the oil humidity at a low level close to the original value. When storing oil and paper together, the temperature of oil titration must be identical to that of oil sampling. Measure and note the oil temperature during sampling directly in the oil; don't use the transformers thermometer.

The transportation time should be short; the time between sampling and measurement ought to be below one week, depending on the tightness of the vessel. Check for the measurement date of the laboratory.

Quality Assurance in the Laboratory

- 1. Before titration the instrument should be conditioned resulting in a stable drift below 12 μ g/min. During this the gas circulation and the oven if available should be active. For external heating three single measurements of the blind value will give a reliable average to compensate for this factor.
- 2. Water standards (liquid for direct injection, solid for external heating) should periodically check the reactivity of the reagent. A high or unstable drift value is insufficient to indicate a consumed reagent.
- 3. The heating temperature for paper should be 160°C, for pressboard 180°C. Thus the reproducibility increases, the measurement time decreases and the sample will not be oxidized.
- 4. To separate oil from cellulose an oil-solving medium such as heptane is much more suitable than methanol.
- 5. For titration of oil the coulometric titration with direct injection seems to be the most reliable method. External heating in an oil chamber cannot be recommended because of possible side reactions in aged oils. Volumetric titration has a lower sensitivity compared to coulometric titration.

7.3 List of Publications

- [1] M. Koch, M. Krüger: "Moisture Determination by Improved On-Site Diagnostics", *TechCon Asia Pacific*, Sydney 2008
- [2] M. Koch, S. Tenbohlen, M. Rösner: "Moisture Ingress in Free Breathing Transformers", *CMD 2008 International Conference on Condition Monitoring and Diagnosis*, Beijing, China, 2008
- [3] M. Koch, M. Krüger: "A Fast and Reliable Dielectric Diagnostic Method to Determine Moisture in Power Transformers", *CMD 2008 International Conference on Condition Monitoring and Diagnosis,* Beijing, China, 2008
- [4] M. Koch, M. Krüger, S. Tenbohlen: "Moderne Verfahren zur Wasserbestimmung in Leistungstransformatoren" Stuttgarter Hochspannungssymposium 2008: Zustandserfassung und -beurteilung von Betriebsmitteln des elektrischen Netzes, Leinfelden, Germany, 2008
- [5] M. Koch, S. Tenbohlen, D. Giselbrecht, C. Homagk, T. Leibfried: "Onsite, Online and Post Mortem Insulation Diagnostics at Power Transformers", *Cigré* SC A2 & D1 Colloquium, Brugge, Belgium 2007
- [6] M. Koch, S. Tenbohlen, M. Krüger, A. Kraetge "Improved Moisture Analysis of Power Transformers Using Dielectric Response Methods" MATPOST, *3rd European Conference on HV & MV Substation Equipment,* Lyon, France, 2007
- [7] M. Koch, S. Tenbohlen: "The Breakdown Voltage of Insulation Oil under the Influence of Humidity, Acidity, Particles and Pressure", International Conference on Advances in Processing, Testing and Application of Dielectric Materials APTADM, Wroclaw, 2007
- [8] M. Koch, S. Tenbohlen: "Systematic Investigations on the Evolution of Water Vapour Bubbles in Oil-Paper-Insulations" *Proceedings of the XVth International Symposium on High Voltage Engineering, ISH*, Ljubljana, Slovenia, 2007
- [9] M. Koch, S. Tenbohlen, J. Blennow, I. Hoehlein: "Reliability and Improvements of Water Titration by the Karl Fischer Technique" *Proceedings of the XVth International Symposium on High Voltage Engineering, ISH*, Ljubljana, Slovenia, 2007
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- [11] M. Koch, S. Tenbohlen: "Der Bubble-Effekt und das Risiko eines dielektrischen Fehlers in Leistungstransformatoren" *ETG Fachtagung "Diagnostik elektrotechnischer Betriebsmittel"*, Fachbericht 104, Kassel, Germany, 2006

- [12] M. Koch, S. Tenbohlen: "Ein neues Verfahren zur Online-Feuchtemessung in Leistungstransformatoren" ETG Fachtagung "Diagnostik elektrotechnischer Betriebsmittel", Fachbericht 104, Kassel, Germany, 2006
- [13] M. Koch, S. Tenbohlen, T. Stirl: "Advanced Online Moisture Measurements in Power Transformers" CMD 2006 International Conference on Condition Monitoring and Diagnosis, Changwon, Korea, 2006
- [14] M. Koch, S. Tenbohlen: "Wasser in Leistungstransformatoren Richtig messen und den Zustand beurteilen" Stuttgarter Hochspannungssymposium 2006: Zustandserfassung und -beurteilung von Betriebsmitteln des elektrischen Netzes, Leinfelden, Germany, 2006
- [15] M. Koch, T. Lange, G. Löhning, H. Schwarz: "Optimierung und Feldberechnung an einer impulsbasierten EMV-Systemprüfanlage" *HF-Report*, September/Oktober 2005, S. 28 – 33, München, Germany, 2005
- [16] M. Koch: "Improved Determination of Moisture in Oil-Paper-Insulations by Specialised Moisture Equilibrium Charts" *Proceedings of the XIVth International Symposium on High Voltage Engineering, ISH*, p. 508, Beijing, China, 2005
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- [20] M. Koch, K. Feser: "Reliability and Influences on Dielectric Diagnostic Methods to Evaluate the Ageing State of Oil-Paper Insulations" *International Conference* on Advances in Processing, Testing and Application of Dielectric Materials APTADM, pp. 95-101, Wroclaw, 2004
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 ür Leistungstransformatoren" ETG Fachtagung "Diagnostik elektrischer Betriebsmittel", Fachbericht 97, S. 265-270, Köln, Germany, 2004
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7.4 Definitions and Abbreviations

Water Vapour Pressure

The partial pressure exerted by water vapour is called the water vapour pressure p. In a mixture of ideal gases the partial pressure of one component is proportional to its concentration in the mixture. Summing up all partial pressures in a mixture gives the total pressure P_T . At the surface of boiling water the saturation water vapour pressure becomes equal to the atmospheric pressure. Water dissolved in oil or cellulose also generates a certain water vapour pressure [Gerrard 1980].

Relative Humidity

Relative humidity *RH* is defined as the ratio of partial water vapour pressure p in a gaseous mixture of air and water to saturation water vapour pressure p_s at a given temperature and pressure. Relative humidity is expressed as a percentage (equation (43)). Capacitive probes and chilled mirror dew point hygrometers measure relative humidity (p. 30). Relative humidity refers to water vapour in gases, whereas water in solids or liquids is described by relative saturation.

$$RH = \frac{p}{p_s} \cdot 100\%$$
(43)

Water Saturation – Relative Saturation

Water saturation or relative saturation *RS* in ideal gases and liquids is the ratio of the actual water content W to the saturation water content W_S . A value of 100 % indicates water saturation whereas zero indicates the total absence of water molecules.

$$RS = \frac{W}{W_S} \cdot 100 \%$$
(44)

Water saturation in *solids* is the ratio of the partial water vapour pressure exerted by the water in the solid in the surrounding gas space under equilibrium conditions to the saturation water vapour pressure.

$$RS = \frac{p}{p_s} \cdot 100 \% \tag{45}$$

Water Activity

The free moisture available in a material is defined as water activity a_w , in opposition to chemically bound moisture [Wiederhold 1997]. It is numerically related to equilibrium relative humidity ERH. However, ERH is expressed in terms of 0-100 % and water activity in 0-1. Water activity represents a very useful assessment of the free moisture of a material for a wide variety of quality purposes, but it does not reflect the total moisture or water content. If water activity is measured in air equilibrated to oil it could be called water-in-oil

activity $a_{W,Oil}$, similarly in equilibrium to cellulose it is water-in-paper activity $a_{W,P}$.

Water Content – Water Relative to Weight

Water mass m_{H2O} in a material related to its weight *m* results in water content *W* or water relative to weight. For moisture in cellulose the measure is per cent of weight (%) and for moisture in oil it is parts per million of weight (ppm). Water content can be related to the mass as sampled m_{as} , or to the dry weight m_d . This thesis uses the latter definition. Commonly Karl Fischer titration (p. 26) determines water mass m_{H2O} . Sometimes also the term "absolute water content" is used meaning water content, although literally a relation is mentioned.

ISO 287 for non-impregnated paper and board:	$W = \frac{m_{as} - m_d}{m_{as}} \cdot 100\%$	(46)
IEC 60814 for oil impregnated paper and board:	$W = \frac{m_{as} - m_d}{m_d} \cdot 100\%$	(47)
	100	

IEC 60814 for oil in ppm:

$$W = \frac{m_{H2O}}{m_{as}} \cdot 1'000'000$$
 (48)

In short, water content refers to weight, whereas water saturation refers to saturation vapour pressure.

Cellulose

In this thesis the word cellulose is used for the transformer materials wood, pressboard and paper. Literally cellulose is a component beside others within the mentioned materials but here for simplification cellulose is used as a collective term.

Dielectric Response or Dielectric Properties?

In English speaking countries it is usual to call a measurement of dielectric properties a "dielectric response measurement", although indeed a superposition of both conductivity and polarisation effects is mentioned. A dielectric measurement at an insulation system always involves conductivity *and* polarization effects. Actually the latter causes the dielectric response in its native sense. Within this thesis the expression "dielectric properties" is preferred to "dielectric response", unless the response is solely mentioned.

a_w	Water activity
$a_{W,Oil}$	Water-in-oil activity
$a_{W,P}$	Water-in-paper activity
С	Capacitance, in F
С'	Real part of capacitance, in F
<i>C</i> "	Loss part of capacitance, in F
C_0	Capacitance as determined by the geometry, in F
C_H	Capacitance between high voltage winding and tank, in F
C_{HL}	Capacitance between high and low voltage winding , in F
C_{HT}	Capacitance between high voltage and tertiary winding, in F
C_L	Capacitance between low voltage winding and tank, in F
C_{LT}	Capacitance between low voltage and tertiary winding, in F
C_T	Capacitance between tertiary winding and tank, in F
D_t	Drift in a titration vessel, in µg/min
D	Electric displacement field
Dira	Analysis software developed within this work
DP	Degree of polymerisation
Ε	Electric field vector, in V/m
E_0	Constant electric field, in V/m
E_a	Activation energy, in eV
ERH	Equilibrium relative humidity, in %
f	Frequency, in Hz
f(t)	Dielectric response function in time domain
$F(\omega)$	Dielectric response function in frequency domain
FD	Frequency domain
FDS	Frequency domain spectroscopy

List of Symbols and Abbreviations

FRA	Frequency response analysis
H_2O	Water
H_2SO_4	Sulfuric acid
Н-СООН	General representation of a carboxylic acid
HV	High voltage
Ι	Current, in A
I_{dep}	Depolarization current, in A
I _{dn}	Normalized depolarisation current, in 1/s
I_{pn}	Normalized polarisation current, in 1/s
I_{pol}	Polarization current, in A
IEC	International Electrotechnical Commission
I_2	Iodine
J	Current density
k	Boltzmann constant; $8,617385 \cdot 10^{-5} \text{ eV} / \text{K}$
KFT	Karl Fischer titration
KOH	Caustic potash solution
LV	Low voltage
m_{as}	Mass as sampled, in g
m_d	Dry weight, in g
<i>m</i> _{<i>H2O</i>}	Mass of water, in g
MODS	Commercial software package to analyse FDS measurements
MV	Middle voltage
N	Ion concentration, in $1/m^3$
OFAF	Forced oil and forced air circulation
ONAN	Natural oil and natural air circulation
OFWF	Forced oil and forced water circulation
Р	Electric polarisation

PDC	Polarisation / Depolarisation Currents
ррт	Parts per million of weight, in µg/g
р	Water vapour pressure, in Pa
p_S	Saturation water vapour pressure, in Pa
P_T	Total pressure, in Pa
R	Ideal gas constant; 8,314472 J / (K mol)
Redia- tool	European research project "Reliable Diagnostics of HV Transformer Insulation for Safety Assurance of Power Transmission System"
RH	Relative humidity, in %
RS	Water saturation or relative saturation, in %
RMS	Root mean square value
RVM	Recovery voltage method
RWE	Rheinisch-Westfälisches Elektrizitätswerk AG
SO_2	Sulfur dioxide
SWRVM	Commercial software package to analyse RV measurements
Т	Kelvin temperature, in K
TAN	Total acid number, in mg KOH/g oil
tan <i>δ</i>	Dissipation factor of a dielectric
TD	Time domain
t	Time, in s
t_C	Charging time, in s
t_d	Discharging time, in s
t _{peak}	Time to peak, in s
U(w)	Sinusoidal voltage, in V
U_{0}	DC voltage, in V
U_C	Charging voltage, in V
$U_{r,max}$	Peak recovery voltage, in V

W	Water content or water relative to weight, in %
W _{IEH}	Water content as measured by the University of Stuttgart, in %
W_L	Water content as measured by an external laboratory, in %
W_S	Saturation water content, in %
Х	Relative thickness of all pressboard barriers and insulation paper in a transformer insulation, in %
Y	Relative width of all spacers of a transformer insulation, in %
$\delta(t)$	Delta-function
ε	Dielectric permittivity
\mathcal{E}_0	Permittivity of vacuum; 8,8541878176 10 ⁻¹² F/m
arepsilon'	Real part of permittivity, reflects polarisability of a dielectric
$\varepsilon^{''}$	Loss part of permittivity, reflects polarisation losses
$\mathcal{E}(\infty)$	High frequency part of permittivity
\mathcal{E}_r	Relative permittivity
$\mathcal{E}_{r,m}$	Measured relative permittivity
9	Temperature, in °C
σ_0	DC conductivity, in S/m
χ	Electromagnetic susceptibility
χ'	Real part of susceptibility, reflects polarisability of a dielectric
χ"	Imaginary part of susceptibility, reflects polarisation losses
Ψ	Water potential
ω	Circular frequency, in 1/s

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