A Review on Chemical Diagnosis Techniques for Transormer Paper Insulation Degradation

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***Abstract*—Energized parts within power transformer are isolated using paper insulation and are immersed in insulating oil. Hence, transformer oil and paper insulation are essential sources to detect incipient and fast developing power transformer faults. Several chemical diagnoses techniques are developed to examine the condition of paper insulation such as degree of polymerization, carbon oxides, furanic compounds and methanol. The principle and limitation of these diagnoses are discussed and compared in this paper.**

***Keywords*— Paper insulation, Degree of Polymeriation, Furan analysis, CO/CO_2 ratio, Methanol**

I. INTRODUCTION

Power transformers represent a vital link in any electrical transmission or distribution network. Unexpected failure won't only cause loss of revenue but it may lead to a catastrophic failure including environmental hazards due to oil spillage. Therefore, it is essential to adopt appropriate monitoring and diagnostic techniques for incipient fault detection to avoid catastrophic failures and help to provide efficient predictive maintenance that improves the reliability of the equipment [1]. Often, power transformer health is referred to the quality of its insulation system which consists of paper insulation immersed in insulating oil [2, 3]. Hence, samples of transformer oil and paper insulation are essential sources to detect incipient and fast developing faults.

Transformer faults generally result from the long term degradation of oil and paper due to the combination of heat (pyrolysis), moisture (hydrolysis) and air (oxidation) [4-6]. Due to electrical and thermal stresses that in-service power transformer experiences, oil and paper decomposition occurs resulting in a number of gases related to the cause and effect of various faults. Gases produced due to oil decompositions are hydrogen (H_2), methane (CH_4), acetylene (C_2H_2), ethylene (C_2H_4) and ethane (C_2H_6), while carbon monoxide (CO) and carbon dioxide (CO_2) are mainly produced by paper decompositions and can be used as a trigger source for paper monitoring [7-9]. The characteristics and concentrations of the gases dissolved in transformer oil vary by the nature of fault, and hence can be used to identify the type of fault. However, the analysis is not always straightforward as there may be more than one fault present at the same time. Transformer internal faults are categorised into thermal or electrical where each fault evolves particular characteristic gases and produces energy from low level to high level of sustained arcing. Partial discharge which produces H_2 and CH_4 is a low level energy fault, whereas arcing that capable to generate all gases including C_2H_2 is considered high level energy fault [7, 9, 10]. Various faults and characteristic gases it produce are illustrated in Fig. 1.

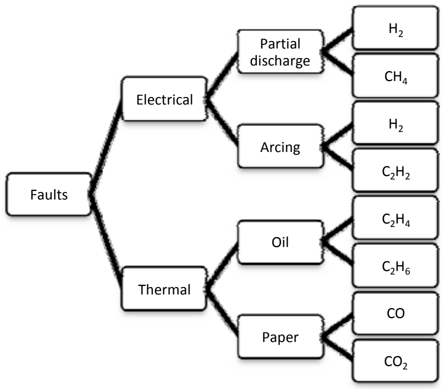

Fig. 1 Types of faults and associated gases [7]

II. PAPER INSULATION

Paper insulation consists of cellulose, hemi-cellulose, lignin and some mineral substance. According to [4], paper insulation is composed of approximately 90% of cellulose, 6-7% hemi-cellulose and 3-4% of lignin, while [5] reports that soft wood Kraft paper consists approximately of 80% cellulose, 12% hemi-cellulose and 8% of lignin and some mineral substances. A dry wood Kraft paper contains 40 to 50% of cellulose, 10-30% hemi-cellulose and about 20-30% lignin [6]. Cellulose is a linear polymer of glucose molecules, which are connected together via glycosidic bond [11]. When degradation of paper insulation occurs, hydrogen bonds are tending to breakdown causing the cellulose molecular chain to be shorter. As a result, some chemical products such as CO, CO_2 and furan derivatives are formed and dissolve in the oil. According to Duval, high rates of paper degradation are indicated when the ratio of CO/CO_2 decreases below 6 [12]. However, the application of CO and CO_2 ratio as an indicator for paper health condition is not reliable due to oil long-term oxidation effect that may produce these gases [5]. To overcome this problem, additional tests such as furan analysis or paper degree of polymerization (DP) is conducted to examine the paper health condition. Lately, due to upgraded thermal paper used in power transformer, which produce less furan derivatives, another possible chemical marker is investigated. According to Jalbert et al.[13] and Annelore Schaut et al.[5] , methanol (MeOH) has the potential to be used as a new indicator to monitor the paper insulation condition. In this paper, several techniques that have been developed in order to determine the condition of paper insulation such as DP, furan analysis, CO/CO_2 ratio and MeOH are presented.

III. PAPER DEGREDATION MONITORING TECHNIQUES

A. Degree of Polymerization (DP)

The degree of polymerization (DP) is a direct technique applied to assess the condition of insulating paper in power transformer as stated in IEC 60450 [14]. DP value reveals a strong correlation between the insulation paper deterioration and formation of aging products. The number of anhydro-β-glucose monomers, $C_6H_{10}O_5$ units (also known as DP) in cellulose chain is a direct indicator of the cellulose decomposition. With DP technique, the length of the cellulose chain is measured by the average DP based on viscosity (DP_V) method to determine the quality of cellulose [6, 15]. Viscometer method to determine DP values was introduced by Staudinger in early 1930's [16] and the correlation of intrinsic viscosity with molecular weight, known as Mark-Houwink equation, was formulated in 1940, [17]. The intrinsic viscosity of a polymer in a dilute solution is correlated to the volume of hydrodynamic sphere of the molecule in solution, which depends on the shape and type of polymer [16]. However, Mark-Houwink equation was only valid for dilute solutions approximately between 0.1 to 1.0%, since the relationship of DP and intrinsic viscosity is linear within this range only [16]. Therefore, the standard ASTM D4243-99 clearly stated that the value of intrinsic viscosity (η) must remain below 1[15]. Huggins-Kraemer [17] proposed a technique to measure η based on the concentration of cellulose (g/100ml of solution). In ASTM D4243 standard procedure, Martin's formula is used to calculate the intrinsic viscosity which is quite similar with Huggins-Kraemer's equation. The first standard procedure to measure the average viscometric of degree of polymerization was published in 1974 kwon as IEC 450 (known later as IEC60450)[14]. Based on IEC 60450, a sample of insulation paper from servicing transformer is required for direct measurement of DP [18]. This paper sample must be taken from locations that have the most rapidly aging paper (hot spot locations) [19]. Emsley et al.[20] developed a first-order kinetic equation that relate the reaction rate at any time with the number of unbroken chain bonds available.

Shroff and Stannett [21] proved that the paper tensile strength is proportional with the DP value until the transformer go to end of life. This result is supported by the study done in [22] as shown in Fig. 2. New Kraft paper has an average length of DP around 1000 to 1500 and the tensile strength is about 1200. When DP value decreases from 1000 to 450, it is considered as moderate deterioration and the strength is virtually constant. However, when DP value is falling down below 450, it is an indicator that the mechanical strength in paper is critical. Paper colour changes to dark brown when DP values is in the range of 200 to 250, and when it reaches a value between 150 to 200, the paper is considered to have no mechanical strength anymore, and the transformer life is over [4-6, 9, 23, 24]. The correlation between DP of insulation paper and its mechanical strength is summarized in Table I.

Gel Permeation proposed a method that relates DP values with the operation temperature. It is reported that the value of DP begins to decrease at a temperature between 120-140°C, and rapidly decreases with the increase in operating temperature. It goes to end of life criteria at 160-180°C [6].

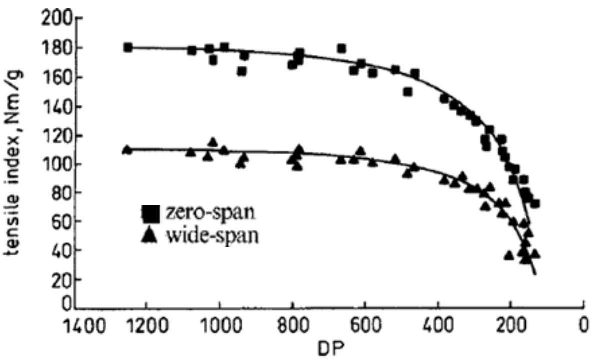

Fig. 2 Tensile strength and DP correlation [22]

Table I Correlation between DP and mechanical strength

DP Value	Mechanical Strength	Significant of Transformer
1000 -1500	Greater (New paper)	Healthy Insulation
450 – 1000	Constant (Normal operation)	Moderate Deterioration
250 - 450	Critical (Lower requirement)	Extensively Deterioration
200 - 250	Nearly Losses Strength	Crucial Deterioration
<200	Zero Strength (Endless)	End of Life Criteria

B. Carbon Oxides Concentration (CO and CO_2)

An indirect technique for paper insulation assessment is by using dissolve gas analysis (DGA). In Opposing to Degree of Polymerization method, DGA can be easily applied for an operating transformer [15, 25]. By analyzing the insulating oil of transformers, specific gas concentrations, its generation rates and total combustible gases can be detected using DGA approaches [26]. Gases dissolved in transformer oil can be extracted using ASTM D3612 – Test Method for Analysis of Gases Dissolved in Electrical Insulating Oil by Gas Chromatography[25] or IEC Standard 567- Guide for the sampling of gases and of oil from Oil-Filled electrical equipment and for the analysis of free and dissolved gases. Tamura et al.[8] reported a relationship between the amount of carbon oxides, CO and CO_2, dissolved in transformer oil and the degree of polymerization of insulating paper. De Pablo [27] reported that water and carbon dioxide are the main by-products of the thermal degradation of cellulose. Hence, the ratio of CO/CO_2 is normally used as an indicator of thermal decomposition of cellulose[28]. According to IEEE Standard C57.104, the ratio of CO/CO_2 is normally more about seven, while the respective values of CO_2 and CO should be greater than 5000 ppm and 500 ppm in order to improve the certainty factor. According to [29], when this ratio is less than 3, a severe paper degradation is expected. When the ratio exceeds 10, it indicates a fault of temperature less than 150°C. According to Duval et al.[30], fault start to arise when the

CO/CO_2 ratio is less than 6, while higher CO/CO_2 ratio is proposed by Kan and Miyamoto [31, 32] after considering the absorption phenomenon of CO_2 and CO into paper insulation. Diagnosing paper insulation condition using CO/CO_2 is not reliable since carbon oxides may be generated from the long-term oxidation of oil components or could present as a result of atmospheric leak [33].

C. *Furan Analysis*

Furanic compounds that are mainly produced due to paper oxidation and hydrolysis processes could be directly extracted from the oil to characterize the thermal decomposition of insulation paper[12]. Furan concentration in transformer oil depends on the mass ratio between oil and cellulose [26]. Levoglucosan leads to the formation of furfural products at temperature above 200°C [19] and the rate of furan production is related to the fractions of glycosidic broken bonds [34].

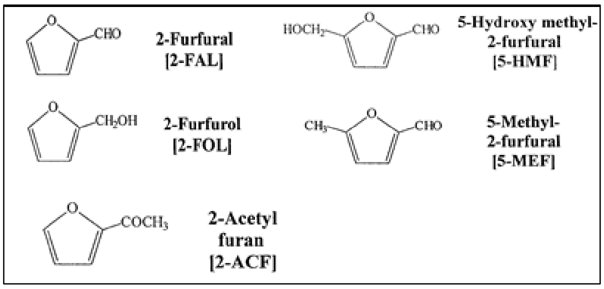

Fig. 3 Furanic compounds detectable in transformer oil [34]

Furanic concentration in oil can be quantified by using High Performance Liquid Chromatography (HPLC) or Gas Chromatography-Mass Spectrometry (GC/MS) based on American Society for Testing and Material (ASTM D5837- Standard Test Method for Furanic Compounds in Electrical Insulating Liquids by HPLC; and ASTM D3612 – Test Method for Analysis of Gases Dissolved in Electrical Insulating Oil by Gas Chromatography) [25, 35]. Both techniques are acknowledged to provide accurate and reliable measurement of furan derivative concentration in transformer oil. Five furan derivatives are related with cellulosic insulation degradation in transformer oil; 2-Fulfural (2FAL), 2-Fulfurol (2FOL), 5-Hydroxy methyl-2-furfural (5HMF), 5-Methyl-2-furfural (5MEF) and 2-Acetyl furan (2ACF) as shown in Fig. 3.

It is reported that the first application of furan analysis to assess thermal degradation is initiated by Central Electricity Generating Board (CEGB) in UK around 1980s [36]. According to Scheirs et al. [34], furanic compounds are dominant by 2FAL concentration during hydrolysis degradation of cellulose, and Nevell has identified 2FAL, 5HMF, and 5MEF as the major products of hydrolytic degradation of cellulose for a temperature range of 100-200°C. Experimental measurements proved that the DP value decreases with the increase of furan concentration in transformer oil and there is a logarithmic relationship between the concentration of 2FAL in the oil and the DP [21, 37, 38]. Advanced investigation by Pahlavanpour et al. [39] reported that only 2FAL and 5HMF are detected at a temperature of 120°C either. The concentration of both products is increased with increasing the temperature until 160°C, after which it starts to decrease. Emsley et al. [37] reported that the concentration of 2FAL is the highest among furan products during accelerated aging test for wood-based paper, cotton-based paper and pure cotton linters. They also reported that the concentration of all furan derivatives increases exponentially with time to a maximum value and then decreases.

Chendong et al. proposed a linear relationship between furfural concentration in logarithmic scale and DP as below [24].

$$\log(2FAL) = 1.51 - 0.0035(DP_v) \quad (1)$$

where 2FAL represented the furfuraldehyde concentration in mg/L. De Pablo [27] reported the following correlation between 2FAL and DP.

$$DP_v = \frac{7100}{8.88 + 2FAL} \quad (2)$$

where 2FAL refer to the furfural concentration expressed in mg/kg of oil. However, it has been noticed that not all winding paper degrade with the same extension, since it depends on the transformer operating conditions. This formula is then revised by Serena [27]. The revised formula is given as:

$$DP_v = \frac{7100}{8.88 + 2FAL + 1} \quad (3)$$

Emsley et al.[37], reported that the rate of change of 2FAL concentration in oil is more important than its absolute level. They found that 2FAL concentration increases significantly when DP is below 400 and expressed the production of 2FAL in terms of the reaction rate constant for the exponential portion by using Arrhenius relationship. The idea was then improved by Cheim et al. [40] and translated into the mathematical equation below.

$$DP_v = \left(\frac{2FAL}{\lambda}\right)^{\frac{1}{\psi d}} \quad (4)$$

where 2FAL is expressed in particle per million (ppm), λ, , ψ and d are constants that depend on the paper type and winding longitudinal temperature gradient[41].

Abu-Siada et al.[4] proposed the use of UV-Vis Spectroscopy to measure furan instead of using either HPLC or GC/MS. They found a strong correlation between furan concentration level and oil spectral response bandwidth and peak absorbance as shown in Fig. 4. Table II summaries the relationships between oil spectral response bandwidth and the corresponding DP level[4].

Table II. Correlation between furan spectral response and DP level [4]

Bandwidth (nm)	DP Value	Significance
300-350	1200-700	Healthy Insulation
350-365	700-450	Moderate Deterioration
365-445	450-250	Extensive Deterioration
>445	<250	End of Life Criteria

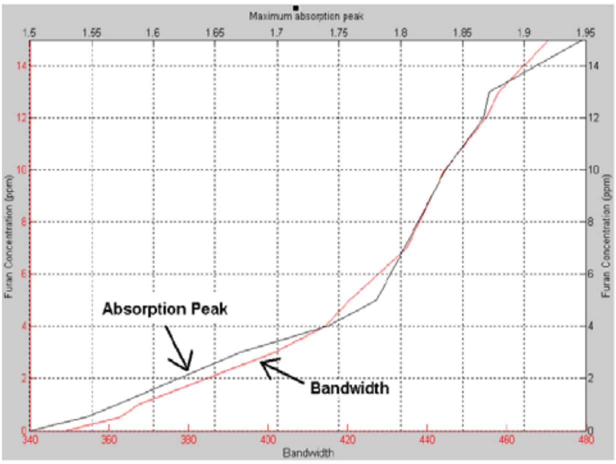

Fig. 4 Correlation between furan concentration in transformer oil and its spectrum response parameters[42]

D. Methanol as a new chemical marker for paper degradation

With the new thermally upgraded paper and the use of vegetable oil as alternative to mineral oil, the use of 2FAL as cellulose insulation degradation indicator could be questionable. Several observations and studies [5, 20, 43-45] show that 2FAL detection for thermally upgraded paper is too low. Field study at Manitoba Hydro [46] shows that some of the failure transformers' oil samples either do not contain 2FAL or the amount of 2FAL is too low, and not detectable.

A few years ago, some researchers investigated the possibility of using Methanol as a chemical degradation marker for paper insulation. From thermal-ageing tests done by Jalbert et al.[13], it was proved that among molecules detected, methanol has the possibility to be used for monitoring paper depolymerization under normal transformer operating conditions. Jalbert et al. [43] reported that 94% of oil samples collected from in service transformer shows the presence of methanol. This results has been supported by further investigation done by Gilbert et al.[33] and Annelore Schaut et al. [5]. Stability and aging tests in [5], proved that MeOH is not an oxidation product, and it is formed as a result of paper degradation. The observation also shows that MeOH is not affected by oil aging condition [47]. Comparative study between MeOH and 2FAL shows that MeOH gives faster indication for early stages of paper degradation rather than 2FAL [5]. Annelore Schaut et al. reported that a linear correlation exists between DP_V and formation of MeOH even at early stages of its formation [5].

Kinetic study of the degradation of thermally upgraded papers in oil conducted by Gilbert et al.[33] has confirmed a strong correlation between MeOH and the rupturing of 1,4-β-glycosidic bonds of cellulose, hence MeOH can be used as cellulose-degradation indicator. In 2012, Jalbert et al. [48] published a standard procedure to analyze of MeOH resulting from cellulose degradation in mineral oils.

Table III summarizes the main advantage/disadvantages of all condition monitoring techniques discussed above for power transformer solid insulation.

Table III. Advantage/disadvantages of various paper diagnosis

Method	Advantages	Disadvantages
CO/CO_2	- Can be easily measured using routine DGA analysis, and can be used as a trigger for further analysis.	-May be resulted from oil at normal temperature due to long term oxidation or due to atmospheric leak.
DP	-Very accurate way to measure the quality of cellulose and paper mechanical strength.	-Impractical to apply for in-service transformer and open breath transformer.
Furan	-Furan level is correlated to DP and mechanical strength of the paper and can be measured through oil analysis.	-Low detection in thermally upgraded paper and vegetable oil; and it depends on the content of manufactured paper.
MeOH	-A linear correlation existed between MeOH and DP from an early stage.	-still in research phase and not fully matured.

IV. CONCLUSION

Among all the chemical diagnosis techniques, DP value is the best to assess the condition of paper insulation in power transformer. However this technique is impractical to be implemented for in-service transformer as it is impractical to acquire paper samples from an operating transformer. Since CO/CO_2 concentration and furan analysis are measured using oil analysis, both chemical diagnoses are widely used in industry for the last three decades. Recently, some studies that were conducted on thermally upgraded paper used in power transformer revealed the potential use of Methanol as a chemical indicator for paper insulation monitoring. However, this finding is still in the research phase and not yet fully matured.

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