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Method for lifetime estimation of power transformer mineral oil

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HIGHLIGHTS

- The paper proposes a simpler and faster method for mineral oil lifetime estimation.
- The proposed method is based on a single ageing test at higher temperature and the activation energy obtained by DSC.
- The obtained results well agree with those derived from the present standardized method.
- The proposed method is effective in terms of costs and experimental time.

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ABSTRACT

Large amounts of mineral oil are used in electrical equipments as insulation and cooling medium. To avoid damages and cut-off of power electricity supply it is necessary to evaluate the mineral oil condition. Lifetime estimation of mineral oil which is based on accelerated thermal ageing at three temperatures (according to IEC 60216-1/2001) requires a long experimental time. This standardized method permits the lifetime calculation using a model based on the equation $\ln D(T) = a + \frac{b}{T}$, where a is the intercept and the slope of the plot $\ln \tau_x$ versus $1/T$, b is a material constant, T is the accelerated ageing temperature and τ_x is the time to reach the acceptable limit value of a chosen degradation parameter. In the present paper, mineral oil lifetime at 80 °C ($D_1 = 1.53 \times 10^5$ h) has been obtained on the basis of accelerated ageing tests at three different temperatures (115 °C, 135 °C and 155 °C) and choosing the oil electrical resistivity (ρ) as degradation parameter. The activation energy of the ageing process determined in these conditions was 101.8 kJ/mol.

To reduce the test duration, this paper proposes a simpler method based on a single ageing test at a higher temperature and the activation energy of the oxidation reaction being determined from non-isothermal Differential Scanning Calorimetry (DSC) measurements. According to the proposed method, the plot ρ versus τ (ageing time) was drawn only for the accelerated ageing temperature T = 155 °C and the lifetime value (τ_x) for this temperature was experimentally obtained. In this case, the activation energy value E_a corresponding to oxidation reaction was determined (E_a = 102.4 kJ/mol). Based on both E_a and the τ_x at 155 °C, the lifetime at 80 °C ($D_2 = 1.86 \times 10^5$ h) was determined and compared with D_1. The differences between D_1 and D_2 values are acceptable and the proposed method is effective in terms of costs and experimental time (the experimental time is reduced by a factor of 12–14) as compared to the present standardized method.

1. Introduction

The power transformers which operate in electric energy distribution and transmission grids have the insulation systems made in cellulose paper and mineral oil. The mineral oil represents 70–80% of the insulation system weight which means, for example, about 80 thousands liters in the case of a 150 MVA transformer [1]. During the operation, both cellulose paper and oil are subjected to electrical, mechanical, thermal and environmental time variable stresses and leading to the worsening of the electrical properties of the mentioned materials. Moreover, there are several power transformers which have been installed more than 30 years ago presenting different ageing levels of their insulation systems; hence they might fail at any time [2]. It is known that the failure

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of a power transformer causes significant expenditures related to its reparation or replacement as well as financial losses compensations required by consumers. Therefore, power transformers users must know as much as possible the remaining lifetime of their equipment in order to take the best decisions concerning the equipment maintenance, refurbishment or replacement [3,4].

Several methods for the diagnosis of the transformers insulation systems, based on the elapsed exploitation time have been proposed. The remaining lifetime can be calculated and an efficient management avoiding the early failures and extending the transformers lifetime (by different maintenance procedures, such as oil refurbishment) can be developed [5–14]. Generally, these methods are based on the analysis and monitoring of some physico-chemical characteristics of both oil and paper subjected to high temperatures, water, oxygen, electrical discharge, vibrations, etc. [11–14]. The estimated lifetime of oil and paper, determined before the transformer start up, are taken also into account by these methods.

Generally it is accepted that heat is the most important stress factor that induces the degradation of an insulation systems during the transformers service. Assuming that the thermal stress is preponderant, the most commonly used method for insulations lifetime assessment is based on accelerated thermal ageing tests at three temperatures (see for example IEC 60216-1/2001 [15]). In case of transformer oil and paper these temperatures range between 110 and 160 °C [16]. Unfortunately, the accelerated thermal ageing method is time-consuming especially for thermal ageing temperatures close to the operation conditions (for example, an ageing test at $T = 115$ °C requires about one year).

It is known that the hydrocarbon compounds (like simple hydrocarbons, mineral oils or hydrocarbon polymers) are unstable under the influence of elevated temperatures, especially, in the presence of oxygen. Generally, the oxygen can degrade the hydrocarbons by processes based on free radicals reactions which generate hydroperoxides [16]. The hydroperoxides are unstable and decompose to other free radicals leading finally to water and ketones as well as to different other oxidation products, such as carboxylic acids alcohols and phenols. Different gaseous (such as CO, CO_2, and low molecular mass hydrocarbons), liquid (e.g. acids, ketones, alcohols, tars and water) and solid phase products (e.g. asphalts and carbenes) were identified as oxidation products of hydrocarbon oils [17]. An oxidation mechanism and discussion related to this topic is presented in Ref. [20].

The presence of these oxidation products, leads to the increase of water absorption and to the worsening of the useful properties of the hydrocarbon based materials [17,18]. In case of transformer mineral oils, the solid phase oxidation products (sludge) may block the oil circulation ways and deposit on the transformer's windings and tank, making difficult the heat transfer [17–19]. The carboxylic acids dissolved in the oil have a negative effect on the paper insulation and conductors of the windings, while the volatilized acids can produce corrosion attack on the top part of the transformer.

This paper presents a simpler and time effective method for lifetime estimation of transformers mineral oil, which assumes that the thermo-oxidative reaction is the preponderant reaction of oil degradation during transformers operation. This method is based on the assessment of the activation energy of oxidation reaction by Differential Scanning Calorimetry (DSC) and a single accelerated thermal stress test at a relatively high temperature (155 °C).

2. Experimental

2.1. Materials

The mineral oil used in this study was of type Mol To 30.01R (produced by Mol Lub Hungary Kft.) which is a mineral oil without antioxidant additives, special manufactured for power transformer insulation systems. The product contains 53.1% paraffinic carbons, 39% naphthenic carbons and 7.1% aromatic carbons and has 2-furfural content lower than 0.1 mg/kg. It was kindly supplied by Mol Romania Petroleum Products® and it was used without any supplementary conditioning.

2.2. Methods

Accelerated ageing experiments were performed at different temperatures, namely $T_1 = 115$ °C, $T_2 = 135$ °C and $T_3 = 155$ °C using an oven with forced air circulation. In view to perform the accelerated thermal ageing, the oil was placed in sealed glass cells. At different time periods, the aged oil samples were taken-out and the absorption/resorption currents were measured using a Keithley 6517 electrometer and a special IRLAB liquids cell.

The oil resistivity (ρ) has been calculated from the measured absorption currents (according to IEC 60247) based on the following expression:

$$\rho = \frac{U_0}{i_a(60)} \frac{S}{d}, \tag{1}$$

Were $U_0 = 300$ V is DC applied voltage, d is the distance between the electrodes of the cell and $S$is the electrodes area. $i_a(60)$ represents the absorption current value measured after 60 s from the initial applied voltage moment [40].

DSC (Differential Scanning Calorimetry) measurements were performed in non-isothermal mode (constant heating rate) on a Setaram 131 EVO apparatus, in the following conditions: sample weight 3.9 ± 0.7 mg, air flow 50 mL/min, aluminum pans of 100 μL capacity, temperature range 30–310 °C and heating rate of 2, 4, 6 or 10 °C/min.

Because the oil was fairly volatile and evaporated before oxidation, the sample containing pans were capped by lids which suppressed the evaporation process. A volume of around 70 μL of air was present in each pan enabling the oxidation process in static atmosphere. An example of a DSC curve is shown in Fig. 1. The oxidation onset temperature (OOT) was calculated, according to ASTM E2009-02 [21] as the intersection point of the recorded baseline and the slope of the oxidation exotherm, using the specific function of Calisto Data Processing (Setaram Inc.) software. The $T_{0.6}$ parameter was calculated as the temperature at the 0.6 of the oxidation peak height (Fig. 1). The thermal effect (ΔH_{ox}) and the peak temperature of exotherm oxidation peak (T_m) were calculated using the integration function.

The activation energy of oil oxidation has been evaluated from DSC measurements, using the onset of the oxidation peak at ~185 °C (Fig. 1). The values of the $T_{0.6}$, or heating rate (β) were

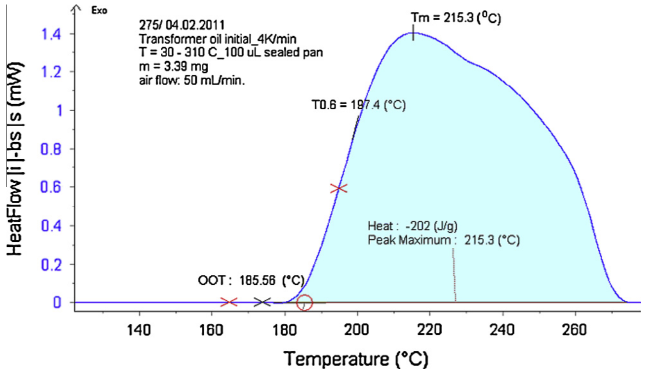

Fig. 1. DSC curve of the mineral oil (heating rate 4 K/min.) and determination of the oxidation kinetic parameters.

Table 1
Fitting parameters of the linear relationships ($y = a' + b'x$) corresponding to different methods and the calculated activation energy E_a in kJ/mol.

Temperature parameter	Method, ref., equation (no.)	Plotting	Corr.	a'	b'	E_a (kJ/mol)	$E_{a,2}$ (average, kJ/mol)
$T_{0.6}$	Kissinger [22,23] $\ln\frac{\beta}{T_f^2} = -\frac{E_a}{RT_f} + C_1$ (2)	$\ln\frac{\beta}{T_f^2} = f(1/T_f)$	0.9997	14.81	−12,096	100.5	102.4
	Ozawa [22,24] $\ln\beta = -1.0518 \cdot \frac{E_a}{RT_f} + C_2$ (3)	$\ln\beta = f(1/T_f)$	0.99978	29.17	−13,080	103.3	
	Boswell [22,25] $\ln\frac{\beta}{T_f} = -\frac{E_a}{RT_f} + C_3$ (4)	$\ln\frac{\beta}{T_f} = f(1/T_f)$	0.99986	22.01	−12,569	104.4	
	Starink [22] $\ln\frac{\beta}{T_f^{1.8}} = -\frac{E_a}{RT_f} + C_4$ (5)	$\ln\frac{\beta}{T_f^{1.8}} = f(1/T_f)$	0.99971	16.23	−12,210	100.5	
T_m	ASTM E698 [29] $\ln\frac{\beta}{T_{\max}^2} = -\frac{E_a}{RT_{\max}} + C_5$ (6)	$\ln\frac{\beta}{T_{\max}^2} = f(1/T_{\max})$	0.9962	19.45	−8795	73	73

plotted as a function of temperature according to the isoconversional methods listed in Table 1. Similar calculations were performed for OOT. T_f from Eqs. (2)–(5) in Table 1, represents the temperature at a fixed amount transformed (in this case $T_{0.6}$), R is the general gas constant, E_a is the activation energy and C_1–C_5 are constants. In Eq. (6) – Table 1, T_m is the temperature that corresponds to the maximum of the oxidation peak.

The plots corresponding to above mentioned equations are straight lines as presented for example in Fig. 2, the corresponding slopes being equal or proportional to $-E_a/R$. The average value of the activation energy $E_{a,2}$ (used in further lifetime estimations) was calculated as the arithmetic average of the calculated E_a values.

3. Results and discussion

Two parallel experiments were performed using the same transformer mineral oil. One consisted in evaluation of the thermal lifetime according to IEC 60216-1/2001. This standardized method will be called below the Method M1. Other is based on determination of E_a from DSC and subsequent lifetime evaluation based on a single ageing experiment at a convenient elevated temperature (in this case, 155 °C). This method proposed in the present work will be called below the Method M2.

According to the standardized method, it is assumed that the thermal degradation is caused by the oxidation reactions whose rate k depends on temperature according to equation:

$$k(T) = A\exp\left(-\frac{E_a}{RT}\right), \tag{7}$$

Choosing a certain electrical property α of the mineral oil, as the resistivity (ρ) or its normalized value r ($r = \rho/\rho_0$, where ρ is the oil resistivity at a certain ageing time τ, and ρ_0 is the initial value of the resistivity at $\tau = 0$), the lifetime D (for the life criterion $\alpha = \alpha_{eol}$), can be obtained from the following equation:

$$\ln D(T) = a + \frac{b}{T}, \tag{8}$$

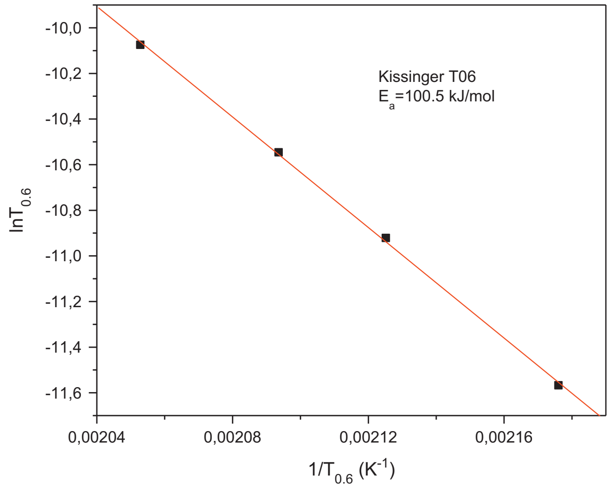

Fig. 2. Evaluation of the mineral oil activation energy according to Kissinger method.

where a is a material constant depending on the limit value of the property chosen as lifetime parameter and the value of the reaction constant $A(\alpha)$, and $b = E_a/R$.

The experimental lifetime determination of the mineral oil, implies to obtain the plots of the α parameter as a function of the ageing time τ, in case of three values of ageing temperatures T_1, T_2, T_3 and graphical determination of the durations τ_1, τ_2 and τ_3, respectively, where τ_i ($i = 1$–3) are the time to reach the selected end-of-life criterion α_{eol} (Fig. 3).

Knowing τ_I values form the three accelerated ageing tests, the parameters a and b can be determined by plotting $\ln\tau_i(T_i)$ versus $1/T_i$, where T_i is the ageing temperature ($i = 1\ldots3$). In these conditions, a is the intercept of the straight line to Oy axis while b is the slope of this straight line. Thus, the lifetime assessment of the mineral oil at a temperature T, in the range of service temperatures (as for example, 80 °C), becomes possible.

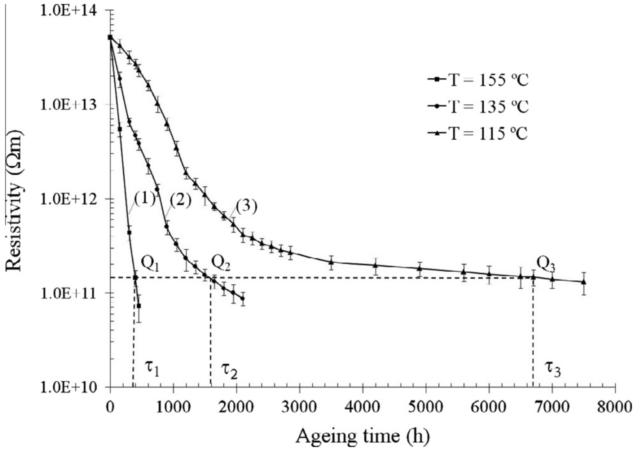

Fig. 4. Variation of resistivity as a function of ageing time at $T_1 = 155$ °C (1), $T_2 = 135$ °C (2) and $T_3 = 115$ °C (3).

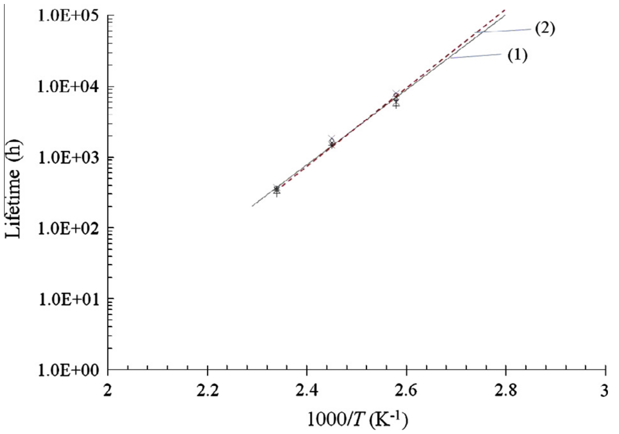

Fig. 5. Mineral oil lifetime curves used M1 method (1) and M2 method (2).

Table 2

The lifetime curves coefficients (a_i and b_i; $i = 1, 2$) and the activation energy E_{ai} values corresponding to the methods M1 and M2 respectively.

Method	Diagnosis factors	a_i	b_i (K^{-1})	E_{ai} (kJ/mol)	D_i (h)
M1	Resistivity	−22.737	12240.24	101.8	1.53×10^5
M2	T_{06}	−22.76	12316.57	102.4	1.86×10^5

Fig. 4 presents the plots of the resistivity ρ (taken as diagnostic parameter) versus the ageing time for three ageing temperatures. In view to reach a good precision, in accordance with IEC 60216-1, each experimental point represented in Fig. 4 has been obtained as an average of five experimental values. It can be seen that, the values of ρ decrease with both the ageing time and ageing temperature, due to the thermally activation of the degradation processes and increase of charge carriers concentration inside the samples. To establish the value of resistivity which can be used as end of life criterion, a preliminary experimental study has been carried out and the resistivity values of several oil samples collected from two power transformers operating since 1977, respectively 1979 were determined. Assuming as end of life criterion, the resistivity value $\rho_{eol} = 1.5 \times 10^{11}\ \Omega$ m, the coordinates τ_i of the Q_i points (Fig. 4), respectively τ_1(350 h), τ_2(1600 h) and τ_3(6700 h) were obtained which enable to plot the lifetime curve (Fig. 5). In the case when all experimental data are taken into account (five resistivity values for each ageing duration) it results fifteen experimental points – five points for each temperature value – which are used in accordance with IEC 60216-1 [15] to obtain lifetime line equation (Fig. 5).

The experimentally found parameters for the Eq. (7), using the coordinate of all fifteen points (Fig. 5) according to the standardized method, are $a_1 = -22.737$, $b_1 = 12240.24$ K^{-1}, and $E_{a1} = 101.83$ kJ/mol (Table 2). Assuming an operation temperature of 80 °C, the lifetime average value $D_1 = 1.53 \times 10^5$ h was obtained. In view to highlight the influence of the ageing curves errors on the estimated lifetime using M1 method, the experimental results shown in Fig. 4 were analyzed and presented in accordance with IEC 60216-1 [15] using simplified procedure. For each ageing temperature, all five resulting points are presented in Fig. 5. The obtained value of the correlation coefficient r is quite high ($r = 0.9942$) and the square root of mean square deviation of points from regression line S_y is low ($S_y = 0.0374$) which means that there is a very good linearity.

A simple examination of the found lifetime values form the experiments performed at T_1, T_2 and T_3 gives an image about the time consumption (and related energy) for such an experiment. Therefore a simpler and more cost and time effective method is to be found.

The kinetic parameters of the oil oxidation process calculated from DSC curves are shown in Table 3. The parameters of the linear fitting of the plots related to different methods for activation energy calculation are presented in Table 1. It can be seen that there is a good fitting on different linear relationships and the resulted E_a values are close enough. The activation energies presented in Table 1 correspond to a same conversion degree, being calculated from $T_{0.6}$. The average value ($E_{a,2} = 102.4$ kJ/mol) of the calculated activation energies has been taken for further lifetime evaluations.

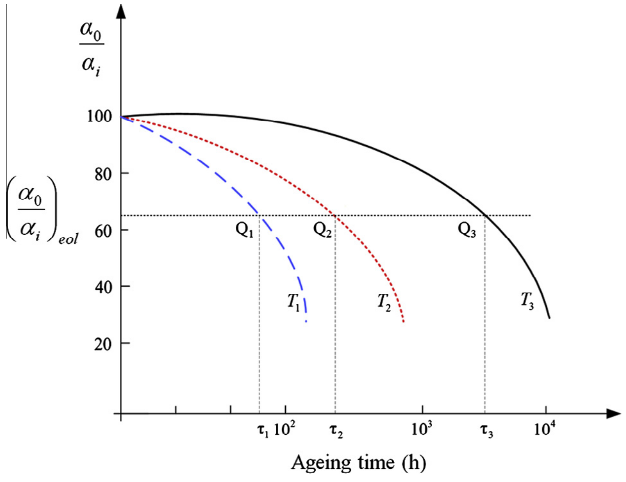

Fig. 3. Accelerated thermal ageing curves and choosing the lifetime curve points.

Comparing the activation energy values experimentally obtained by M1 method ($E_{a1} = 101.8$ kJ/mol) with the value obtained by M2 method ($E_{a2} = 102.4$ kJ/mol), it can be noted that, low differences are obtained (less than 0.6%) using both methods. It is largely recognized that the oil oxidation is an autocatalytic process occurring through a free radicals mechanism. As a consequence, the activation energy is strongly dependent on the oxidative conversion; hence the value of the activation energy chosen for durability evaluation according to method M2 is of major importance. As for example, in our case E_a from OOT has an average value of 139 kJ/mol, while with T_m, E_a is 73 kJ/mol. A higher value of E_a, such as that calculated from OOT, will result in sub-evaluation of the lifetime, while a too small E_a value (e.g. from T_m) will lead to a supra-evaluated durability. Correlating different values

Table 3
Kinetic parameters of the low temperature oxidation process calculated from the non-isothermal DSC curves.

β (K/min.)	OOT (°C)	$T_{0.6}$ (°C)	T_m (°C)
2	177.6	186.4	195.3 212.6
4	185.6	197.4	214.6 230.1
6	190.5	204.5	222.7 265.0
10	196.4	214.0	241.3 276.6

of activation energy E_a (obtained for different values of the temperature corresponding to ascending part of oxidation peak (T_{02}, T_{04}, T_{05}, etc.)) with those determined by method M1, it has been found that T_{06} leads to the activation energy values nearest to that resulting from M1 method. A justification for selection of T_{06} as parameter for E_{a2} calculation is provided by FTIR data: the infrared absorptions in both the regions of 1700 cm^{-1}(carbonyl groups) and 3400 cm^{-1}(hydroxyl groups) are comparable for the oil sample heated up to $T_{0.6}$ and the aged one (according to M1 procedure) near to its lifetime. Hence, at the limit fixed for the resistivity, the oil presented a rather small degradation level similar to that of the sample oxidized in DSC up to $T_{0.6}$ as shown the ATR/FTIR spectra in Fig. 6. The oxidative transformation degrees are rather low in both cases, which mean that the degradation process of mineral oil is still in an early stage. It is perhaps interesting also to observe that the E_{a2} value calculated from T_{06} fails very close to the mean of E_a values calculated from OOT (when oxidation starts) and T_m (when the oxidation process reach a maximum).

With the E_{a2} value from DSC and the D (155 °C) from the accelerated thermal ageing at 155 °C, the parameters $a_2 = -22.76$ and $b_2 = 12316.57\ \text{K}^{-1}$ can be calculated and the lifetime curve can be plotted (Fig. 5). In the case of operation at the constant temperature $T = 80$ °C, the lifetime average value $D_2 = 1.86 \times 10^5$ h was obtained. On the other hand, the influence of the ageing curve errors on the lifetime values predicted by method M2 is smaller because only the Q_1 point coordinates change (Fig. 5). If we consider the same operating temperature $T = 80$ °C, for all five resulting points (Fig. 5) it results that the lifetime value is in the range 1.82×10^5–2×10^5 h (which means an error of ±2.3% compared to average value 1.86×10^5 h). It results that D_2 well agree with D_1, both values having a same order of magnitude; the difference is less than 20%.

The use of a constant value for the activation energy in the case of the proposed method may appear as a too big approximation. As it is known, the degradation process of a material is a sum of multiple concurrent and consecutive chemical reactions (each presenting its specific kinetic parameters) leading to the worsening of the material properties. However, the degradation itself can be seen as a single reaction leading from the initial material M to a degraded one Ma, through the changes in the values of a critical parameter describing the ageing state. The curves of α values versus ageing time present usually simple shapes and the degradation level at the end of life ($\alpha = \alpha_0$) is basically the same, irrespective of the ageing temperature. Besides an electrical property (as the resistivity), the critical parameter can be a chemical one as for example, the oxidation induction time (OIT), the oxidation onset temperature (OOT) or $T_{0.6}$ as considered above. Hence, the degradation process can be characterized by kinetics treatments similar to those applied for simple chemical reactions even their chemical meaning is not precisely defined. Thus, kinetic parameters, such as the activation energy of the ageing process, gain a sense being related to the "reaction" of the critical parameter even the mechanism of this process is considerably more complex than in the case of a simple chemical reaction. On the other hand, it is known that the activation energy of a specific reaction can change during chemical transformations depending on the conversion degree [26,27]. However, in the case of lifetime assessments, different proposed methods [28,30–32], as well as the present proposed method, use constant values for the activation energy (experimentally obtained). A constant value of the activation energy appears to

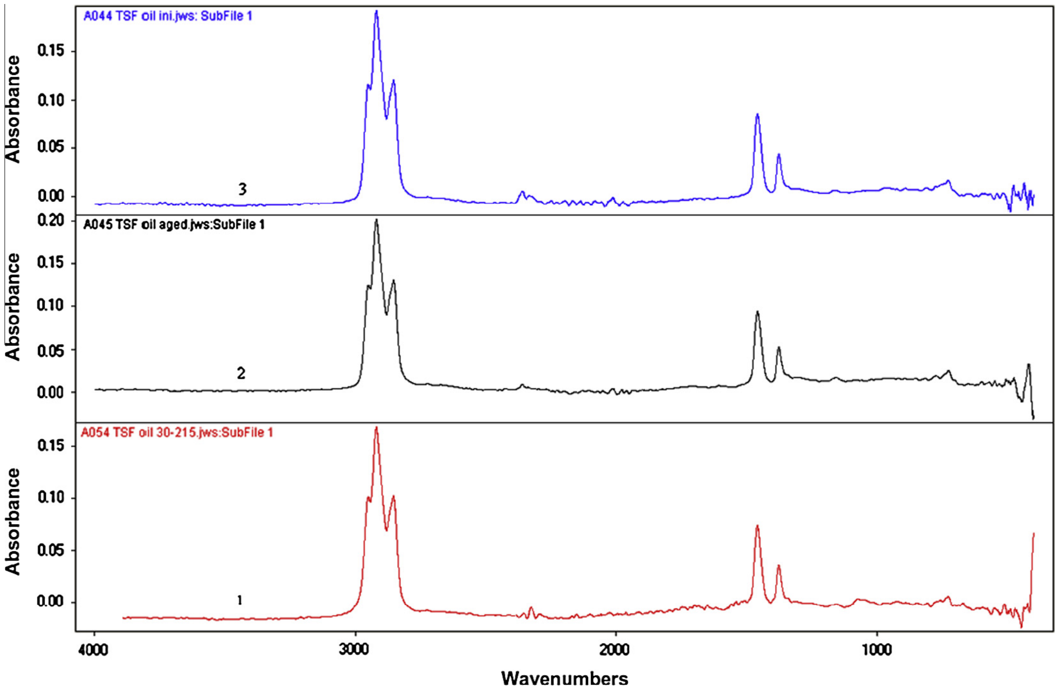

Fig. 6. ATR–FTIR spectra of the transformer oil in different states: (1) initial, as received; (2) after thermal ageing at 155 °C; (3) heated from 30 °C up to 215 °C ($\approx T_{0.6}$).

be justified in this case also by the fact that there is a similar conversion degree of oxidation either for thermally aged sample (at the end of life) or at $T_{0.6}$ in the rapid DSC measurements as mentioned above. Hence, the oxidation of the hydrocarbon components can be globally assumed to occur with a constant rate at a given temperature, for a specific range of conversion (ageing) degrees, in spite of the autocatalytic character of the oxidation process. By setting a specific end-of-life criterion, the material is considered to be aged and its replacement is required, but it still has a certain chemical remaining lifetime (the degradation is not complete from the chemical point of view, as in the case of T_m). Obviously, this level corresponds to a certain chemical composition of the aged material, hence to a certain oxidative conversion degree which is the same regardless the way (the temperature) it is reached.

Lifetime estimation by method M1 required three accelerated ageing tests at 115, 135 and 155 °C using three ovens simultaneously for 350 h (155 °C), 1700 h (135 °C) and 6700 h (115 °C). Also, taking into consideration the required time for samples preparation and diagnosis factor measurement, the total necessary time for mineral oil lifetime estimation exceeds 7000 h. In case of the M2 method, the accelerated thermal ageing test at the highest temperature (155 °C) was used, which consumed around 350 h. Taking into account the time for the samples preparation (including the drying process) and the DSC measurements, a total time of less than 600 h was necessary. Thus, using the method M2, the experimental time was reduced by a factor of 12–14. On the other hand, in case of the second method the costs (energy, labor, samples, etc.) are highly reduced as well. Therefore, it can be said that the second method M2 is time and cost effective and is applicable at least when a rapid lifetime evaluation is necessary. This proposed method can be useful to manufacturers and users of large power transformers for rapid characterization of new oils which can be used both for new transformers and for those which require insulation system restoration (oil refilling and/or paper replacing).

As M2 method seems to be very convenient, it must keep in mind that the activation energy obtained from DSC as well as from accelerated thermal tests did not take into account other degradation mechanisms which occur in real ageing process in-service transformer oil as for example, the presence and the influence of water (appeared as a result of paper degradation and from the environment) [33–35], metals (cooper, aluminum, iron, etc. [36,37] and electric field (partial discharge, electric arcs, space charge resulted by oil electrification phenomena) [38,39]. Depending on the real operation conditions, these factors can lead to different activation energy values. However, such approximation is made also in different other cases, including the standardized methods, as for example those based on measurements of kinematic viscosity [13,40], thermal ageing in the oven [16,41] on laboratory aged samples which do not take into account all the involved stress factors, being commonly accepted that the thermo-oxidation is the preponderant degradation process. Nevertheless, these factors can themselves modify thermo-oxidation activation energy values [34] and our future work will be devoted to this topics.

4. Conclusions

Accelerated thermal ageing standard method (M1) requires long times for lifetime curves obtaining, especially in case of ageing temperature values close to the operation temperature values of mineral oil (up to one year). The assessment of activation energy by the proposed method (M2), allowed the reduction of test duration by a factor of 12–14. The values of the activation energy obtained by both methods are very close, and the evaluated lifetimes are in good agreement. A difference which is less than 18% is still observed, but it is acceptable from practical point of view in the case of long term evaluations.

$T_{0.6}$ parameter of the oxidation peak appears the most adequate to be used for E_a calculation with method M2. The applicability of the proposed method on different oils and a more accurate estimation of oil activation energy, in the presence of both metals (Cu, Fe, etc.) and paper, based on thermal and electrical ageing tests, is ongoing and will be presented in a future paper.

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