Electro-insulating Paper Degradation in Various Electro-insulating Fluids

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The durability and safe operation of electrical equipment and devices with mixed insulation systems (solid/ fluid - electro-insulating paper/oil) is determined by the insulation aging under simultaneous and synergic actions of electrical, thermal and chemical stress factors etc. In this context, degradations of insulating paper exposed to thermal aging in 5 different types of electro-insulating fluid have been studied experimentally. Liquid chromatography determinations have shown that the total content in furan products (resulting from cellulose degradation) in mineral oils is substantially higher than in electro-insulating fluid sorts based on of synthetic ester and/or natural ester (vegetable oil). This is due to the temperature between 90°C and 130°C when the activation energy of the furans formation process is up to 7.5 times lower in mineral oils than in ester-based oils. Degree determinations of cellulose polymerization (viscosimetric method) before and after exposure to heat treatment indicated that mineral oils degrade the electro-insulating paper much more strongly than ester-based oils (both synthetic and natural). Obtained results by liquid chromatography and by viscosity are in accordance with the images obtained by optical microscopy (at X 100).

Keywords: insulating paper, insulation ageing, furan products, degree of polymerization, mineral oils, synthetic esters, natural esters

In sustainable development perspective, the issue of secure power supply of electricity, continuous, at required qualitative parameters and having a minimal negative impact on the environment is of particular importance [1].

Safety in the operation of power electricity transmission/ transport and distribution networks is largely determined by the integrity of solid/liquid mixed insulation (paper / electro-insulating fluid) used at a series of *key* equipment such as transformers, solenoids for null treatment, capacitors, etc.

During the operation of these equipments, mixed insulations - paper/electro-insulating fluid, are exposed to the simultaneous action of some request (stress factors) electrical, thermal, chemical and mechanical.

Under the synergic action of the stress factors, the insulation ages which leads to the failure of the respective equipments - in extreme situations to the explosion and/or incineration (fig. 1 and 2) followed by the dispersion of persistent organic noxes [4, 5] with major environmental effects.

Mixed insulation aging, (paper/electro-insulating fluid) is the consequence of chemical degradations that occur between cellulosic electro-insulating paper, electro insulating fluid, dissolved oxygen in oil, metallic materials used in the manufacture of equipment (especially copper) etc.

Electro-insulating fluid - *transformer oil*- under the action of thermal stress suffers a series of decomposition processes radical (breakage of carbon-carbon bonds and/ or carbon-hydrogen bonds with free radical formation) with formation of flammable gases [6-10] as H_2, CH_4, C_2H_4, C_2H_6, etc., but also CO_2 and CO following reactions with dissolved oxygen - which substantially increases the risk of explosion and/or arson of equipment.

Electro-insulating fluids decomposition is substantially accelerated by contact with copper from equipment that catalyses decomposition processes [10-13] under the action of fast electrons generated by electrical stress (partial discharge and corona effect) [7, 14, 15].

Fig. 1. Washington DC (USA) 2011– from an disabled transformer about 21.000 liters of mineral oil was spilled on the soil and in Potomac River [2]

Fig. 2. Explosion and burning of several transformers in Boto°ani - RO [3] (2017)

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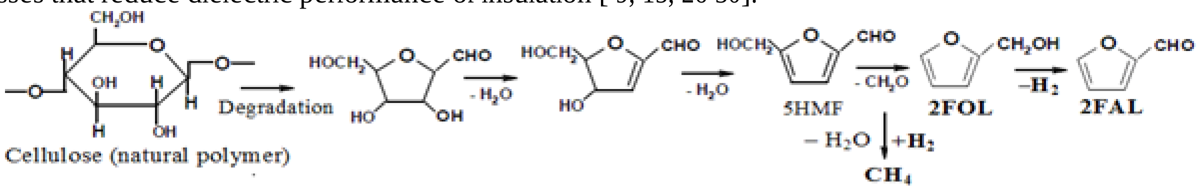
Electro-insulating fluid heating in electrical equipment is the result (depending on the type of equipment) of the Joule effect, film effect, dielectric losses (tgδ) and magnetic losses. In these conditions, the thermal stress is the result of electrical stress characterized by working voltage, currents and frequency. As a result of technical progress, in the last decades a number of highly nonlinear consumers (such as LEDs, switching power supply, including computers, etc.) generate reactive and harmonic powers [16-21], which has led to enhancing film effect and of losses in the dielectric. Another consequence is the partial increase of discharge and the crown effect, which implies global electrical and thermal stress intensification (especially in electro-insulating fluids with magnetic nanoparticles in suspension [22-23]).

Under the influence of thermal stress, the components of the electro-insulating fluids undergo a series of thermo-oxidative processes (with dissolved oxygen participation) which start with peroxide products formation and continue through successive oxidation processes [24, 25] after which increase the viscosity, acidity and water content with consequences (decrease) on permittivity and dielectric rigidity of oils.

Under the action of thermal stress, paper and electro-insulating fluids in mixed insulation systems (solid/liquid) suffer degradation processes (1) with furanic products formation (the majority with negative impact on the environment), processes that reduce dielectric performance of insulation [9, 15, 26-30].



(1)

It is noted that oil dispersed cupper sulphide [31, 32] as well as lower (ethyl and methyl) [33, 34] alcohols formed by paper degradation processes as well as oil thermooxidation have an important role in the insulation of paper degradation.

Numerous recent studies [9, 35-50] have highlighted that gas forming processes and paper degradation processes are much slower in ester-based electro-insulating fluids than in traditional mineral oils used in transformers, which confirms that the use of vegetable oils in the electrical equipment is an environmentally friendly alternative by that ensures sustainability and increased in operational safety [51-54].

The thermal degradation processes of the electro-insulating paper decrease the degree of cellulose polymerization - a parameter determined by IEC 60450 [55]. The method is destructive, as such can not be used for diagnose insulation systems of transformers in exploatation. The degree assessment of aging of the insulation in service and the estimation of the remaining lifetime can be achieved by determining the furan products content and / or in gases [38, 56 - 67] of the oils in exploatation. The degree assessment of aging of the insulations in exploatation and the of the remaining lifetime estimation can be achieved by determining the content in furan products and/or in gases [38, 56 - 67] of the oils in exploatation.

In view of these considerations, the purpose of the paper consist in the comparative study of the thermal degradation of the electro-insulating paper in contact with various sorts of electro-insulating fluids.

Experimental part

Materials and methods

The comparative study regarding thermal aging of insulation paper of Kraft type (Weidmann 22HCC grade) was investigated in five sorts of electro-insulating fluids (table 1, two of which are of mineral origin, one of predominantly vegetable oil, one synthetic ester-based and one vegetable oil with high oleic content (experimentally - *demonstrator model*).

About 200 g of electro-insulating oil sample (table 1) together with about 1dm^2 Kraft insulation paper (Weidmann type 22HCC) were introduce in Erlenmeyer flasks and exposed to thermal aging (under tight conditions - without access to atmospheric oxygen) by storing at 90 ± 3 °C, 110 ± 3 °C and 130 °C ± 3 ^{o}C for 1000 h in a France Etuve XL 980 oven.

By liquid chromatography, using HPLC equipment 1100, manufactured by Shimadzu Corporation Kyoto Japan, determined the content in the furan products of the investigated oils before and after heat treatment. Also, by the viscozimetric method according to IEC 60450 [55], the degree of cellulose polymerisation of the electro insulating paper was determined using an Ubbelohde capillary viscometer produced by COMECTA Spain.

Optical microscopy was carried out with a MM-KKE-M-C-U microscope and a camera SP10 (magnification X 100) provided by HYDAC FILTER SYSTEMS GmbH, Germany and to capture images of the appearance of paper samples before and after thermal treatment in the investigated oils.

Results and discussions

Evolution of the speed of furan products formation based on temperature

The evolution of the formation velocity *k* (the reaction rate constant value) of the furan products (total 5-HMF, 2-FOL, 2-FAL, 2-ACF and 5-MEF results after 1000 h of thermal treatment) is shown in figure 3.

From the analysis figure 3 it can be see that evolution *lnk* function of *1/T* are approximately linear. Applying Arhenius' relationship (2):

$$k = \frac{dC}{dt} = A \cdot e^{-Wa/RT} \quad (2)$$

in which dc / dt represents the total concentration of formed furan products reported at 1000 h of thermal treatment, *k* is the reaction rate constant value, *A* - velocity constant, *Wa* - the activation energy of the processes of formation of the furan derivatives, *R* - the gas constant and *T* - the absolute temperature [*K*], it is obtained that the slope of

Table 1
INVESTIGATED ELECTRO-INSULATING FLUIDS

Sample Code	Composition	Type / manufacturer
Oil 1	Mineral oil	MOL TO-30.01R [68]
Oil 2	Vegetable ester predominantly	Biotemp® [69]
Oil 3	Synthetic ester	LUMINOL [70]
Oil 4	Vegetable ester with high oleic content	MF-UPMEE;P1MF [71]
Oil 5	Mineral oil	Nynas [72]

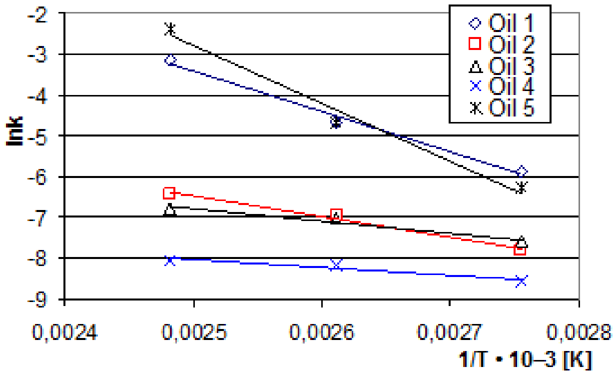


Fig. 3. The evolution of the furans formation velocity depending on temperature

the straight lines (3), lnk is plotted versus the reciprocal of the absolute temperature.

$$\ln k = \ln A - Wa / RT \qquad (3)$$

is (4):

$$Wa / RT = -T \cdot \ln k \qquad (4)$$

From the relationship (4), it is found that the activation energy *Wa* of the furans formation processes at a given temperature *T* increases at increasing *T lnk* - respective decreases the stability of the electro-insulating paper in sample oil.

Decrease evolution of the degree of polymerization of the electro-insulating paper based on temperature

The stability of the electro-insulating paper in the investigated oils was also evaluated by determining the degree of polymerisation of the cellulose DP [55]. The average results of three initial determinations and after 1000 h exposure in the investigated oils are shown in figure 4 where in k_1 represents the velocity of decrease of DP (initial DP - final DP raported at 1000 h exposure into oil at *T* temperature).

In order to compare the compatibility of the investigated oils with the insulating paper, table 2 summarizes the representative data, namely the activating energies reported at *R* and the square deviation from linearity R^2 of the functions described by the relationship (3) obtained after 1000 h of thermal treatment.

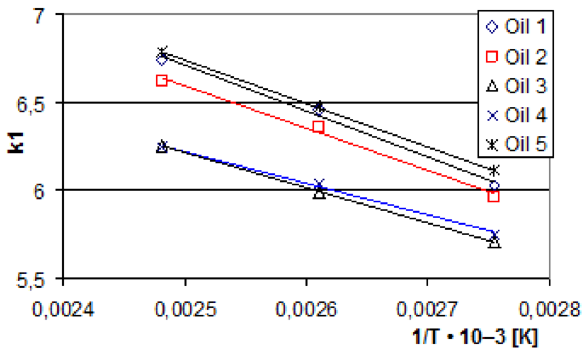


Fig. 4. Decrease evolution of the degree of polymerization of the electro-insulating paper depending on temperature

By analyzing the data presented in table 2 it is found that at temperatures between 90 and 130°C the square deviations from linearity are 0.9415 < R^2 <1.000 and that in the mineral oils Oil 1 and Oil 5, the activating energy of the forming processes of furan products are substantially smaller (up to about 7 times more negative) than in natural esters-based oils (Oil 2 and Oil 4) and synthetic esters Oil 3. It is also found that the energies of activating the breakdown processes of the cellulose polymer chains in the mineral oils are significantly lower than in the ester-based oils. These findings indicate a compatibility with the superior net electro-insulating paper of ester-based oils relative to mineral oils.

These findings are in according with the data reported in [73, 74] and are supported by the macroscopic image (fig. 5) and microscopic (fig. 6) of the paper samples before and after 1000 h of thermal treatment in investigated oils.

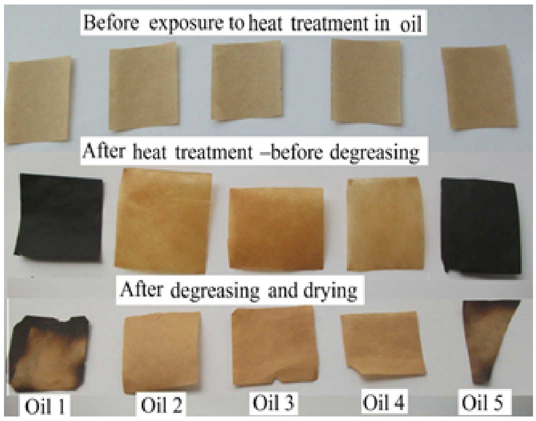


Fig. 5. The image of paper samples exposed to thermal treatment of 1000 hours in oil at 130°C

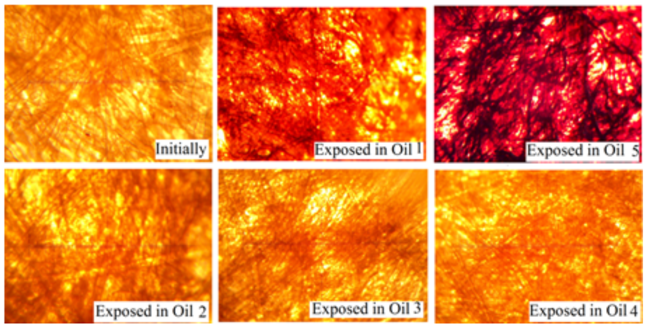


Fig. 6. Microscopic image of paper samples exposed to thermal treatment of 1000 hours at 130°C in investigated oils (X: 100)

By comparison analysis of the images in figure 5 and 6 it is found that the electro-insulating paper - at the level of cellulose fibers - undergoes major changes (degradations up to carbonization of the fiber) in mineral oils Oil 1 and Oil 2, as opposed to ester-based oils, at which microscopic no changes are observed.

Based on the experimental results obtained, it is found that the electro-insulating fluids based on vegetable esters in the applications in electrical equipment in which the fluid is in contact with insulation based on natural cellulose (paper, cotton), compared to traditional mineral oils, have

Table 2

ln k = f (1/T) SLOPES AND SQUARE DEVIATIONS FROM LINEARITY R^2 IN THE INVESTIGATED OILS

Oil sample	Furans formation		Decrease degree of paper polymerization	
	$T \cdot lnk = W_a/R$	R^2	$T \cdot lnk_1 = W_a/R$	R^2
Oil 1	-9.956	0.9904	-2629	0.9920
Oil 2	-5.019	0.9880	-2398	0.9914
Oil 3	-2.969	0.9552	-2011	1.0000
Oil 4	-1.921	0.9415	-1802	0.9954
Oil 5	-14.238	0.9825	-2475	0.9996

the advantage of good compatibility than cellulose, respectively provides thermal stability up to 10 bigger times the insulation.

Thus the durability and safety of the equipment increases accordingly when using vegetable ester-based electro-insulating fluids.

In view of this reason, as well as the fact that natural esters (vegetable oils) are obtained from renewable resources by environment-friendly technologies, they are not toxic and do not release toxic products during their exploatation (furan compounds), are quickly biodegrade in natural conditions, etc., in the perspective of sustainable development and environmental protection [1], it is advisable to replace traditional mineral oils with natural esters (vegetable oils) in electrical applications.

In view of these considerations, it is considered necessary that, through sustained environmental information [75-77], manufacturers and users of electrical equipment should be convinced to abandon the use of traditional mineral oils in favor of vegetable oils.

Conclusions

By liquid chromatography and viscosimetry (to determine the degree of paper polymerization) and optical microscopy, were studies the degradation processes of thermal electro-insulation paper exposed to thermal aging at 90 ± 3 °C, 110 ± 3 °C and 130 ± 3 °C for 1000 h.

Following the processing of the obtained experimental results it was found that after thermal aging:

- furane content of mineral oils increases substantially more (up to 50 times) than in the case of investigated ester electro-insulating fluids - fact which is explained by the activating energies of the furans forming processes up to 7.5 times higher in ester-based fluids than at the mineral oils investigated;
- the degree of polymerisation of the electro-insulating paper decreases as a result of thermal aging - decreases being more pronounced (double) in mineral oils than in investigated ester fluids, in correlation with the activating energies obtained by processing the experimental data;
- images obtained by optical microscopy (X 100) highlight the pronounced structural degradation of thermal treated paper (up to carbonization of the cellulose fiber) in the mineral oils - microscopically there were no visible changes in the paper exposed in the electro-insulating fluids based on the investigations discussed.

These findings lead to the conclusion that the durability and safety in exploatation of the combined insulation systems in electrical equipment is substantially higher (about 10 times) in the case of electro-insulating fluids based on ester than in traditional mineral oils.

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