POWER SYSTEMS AND ELECTRIC NETWORKS

DIELECTRIC LIQUIDS: PAST, PRESENT, FUTURE (REVIEW)

M. N. Lyutikova,1 A. V. Ridel,2 and A. A. Konovalov2

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PJSC "ROSSETI," Moscow, Russia. The paper offers a review of the studies of dielectric properties, as well as aspects of development and application of various insulating fluids used as dielectrics in high-voltage transformers. Transformer oil has been used as a liquid dielectric in high-voltage equipment for more than 130 years. However, transformer oil is a non-renewable natural resource, has low chemical stability, low ignition temperatures, and cannot be decomposed by bioorganisms to yield safer compounds. All this has inspired a search for a dielectric fluid, which would possess the best insulation, environmental, and other performance characteristics. Currently, the focus of electrical engineering companies is gradually shifting toward such alternative biofluids as natural and synthetic compound esters. So far, their application has been limited to certain types of equipment, however, their insulation properties are being actively explored. Recently, researchers have also been studying the electrical insulation properties of nanofluids.

Keywords: mineral dielectric oil; polychlorinated hydrocarbons; synthetic compound ester; plant oils; natural compound ester; nanofluids.

A power transformer is an important component of the electrical grid, the proper functionality of which affects the reliability of supplying power to consumers. The functionality of a high-voltage transformer is largely determined by the condition of its insulation system. For many years, a hybrid system consisting of both paper and liquid insulation has been used in high-power transformers. The paper-oil insulation of the transformer has high dielectric characteristics (e.g., short-term dielectric strength of $100 - 250 \text{ kV/mm}$ (DC voltage) and $50 - 120$ kV/mm (AC voltage)), which ensure its reliable operation [1].

The primary purpose of a dielectric liquid is not only to provide insulation and effective cooling of the current-conducting parts of a high-voltage equipment, but also to protect the cellulose from excessive moisture. To ensure an impeccable performance of its functions, any dielectric liquid must have high dielectric strength, good thermal conductivity, proper chemical and thermal stability, lowest possible flammability and toxicity, adequate viscosity suitable for cold conditions, as well as have the ability to maintain its fundamental properties over an extended period of operation at elevated temperatures and high electrical stresses [2].

In the quest for a dielectric that would meet numerous requirements, various insulation fluids have been developed and tested over 130 years of operating high-voltage oil-filled equipment. Such fluids include: mineral dielectric oils, nonflammable synthetic liquids (e.g., ascarels or polychlorinated biphenyls (PCBs)), plant-based oils, silicone fluids, synthetic and natural esters, and nanofluids. A brief schematic illustrating progress in the development and application of dielectric liquids in various high-voltage equipment is shown in Fig. 1.

Currently, the following dynamically growing areas should be highlighted, which include the creation and analysis of the properties of biodegradable natural and synthetic esters, dielectric liquids with nanoparticles, as well as mixtures of esters with mineral oils.

Since the production of the first high-voltage transformer in 1890, *mineral oil* has been and remains the most widely used dielectric liquid. This oil is a product of petroleum processing obtained by its fractional distillation at 300 to 400°C [3]. Depending on the content of the main classes of hydrocarbon compounds, mineral oils are conventionally divided into paraffinic, naphthenic, and aromatic (Table 1).

The very first mineral oil was produced from paraffinic crudes and contained a large amount of paraffinic hydrocarbons. Such oils had a relatively high chilling point, which

¹ Novosibirsk State Technical University, Novosibirsk, Russia; e-mail: m.lyutikova@mail.ru

² Novosibirsk State Technical University, Novosibirsk, Russia.

complicated their use in high-voltage equipment at low temperatures. The presence of varying amounts of unsaturated hydrocarbons (HCs) made the oil susceptible to rapid oxidation, resulting in the formation of wax deposits, which decrease the effective heat dissipation from the current-conducting parts of the transformer [4].

To replace paraffinic oils, it was suggested to use mineral oils with a somewhat higher content of naphthenic hydrocarbons. Unlike paraffinic oils, naphthenic oils were characterized by low chilling points and exhibited greater resistance to oxidation. Furthermore, naphthene-based dielectric oil prevents the formation of heavy high molecular weight compounds (HMWCs) and keeps them from getting deposited on the windings and elements of the cooling system, as well as from entering the oil channels. This helped avoid local overheating of the transformer winding and prevent the insulation system from exceeding the operating temperature, which consequently resulted in the extended service life of the transformer as a whole [5].

Similar to paraffinic oils, mineral oils with high content of aromatic hydrocarbons, which have been used in highvoltage equipment (e.g., grades TKp and T-750 in Russia) for several decades, are prone to the formation of insoluble residues. A positive aspect of using aromatic oils has to do with their ability to absorb gases formed as a result of thermal and electrical decomposition of hydrocarbon compounds [6].

Over the past 50 years, technologies for producing mineral dielectric oils have improved significantly, which helped minimize the issues associated with high content of unsaturated compounds, low resistance to oxidation, and residue formation. As a result, mineral oils now possess characteristics that satisfy current requirements to the maximum degree possible. At the same time, largely due to environmental considerations, the production of gas-absorbing aromatic oils (e.g., grades TKp and T-750 in Russia) has been discontinued. Currently, paraffinic and naphthenic transformer oils are the ones that are being manufactured on an industrial scale worldwide (Table 2).

Fig. 1. Timeline of the development and application of dielectric liquids in high-voltage equipment.

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Overall, mineral oils effectively perform their main functions of providing insulation and ensuring heat removal. However, their low flame resistance has always been a concern, especially when these oils are used in transformers operating inside buildings and structures (e.g., industrial and commercial complexes, subways, tunnels, fully enclosed or underground substations, etc.). This prompted the industry to search for liquid alternatives to mineral oils, which would pose a lesser fire hazard.

Active research efforts aimed at finding non-flammable dielectric liquids have led to the creation of a number of *synthetic dielectric liquids*, which belong to substituted hydrocarbons, such as fluorinated hydrocarbons, chlorinated diphenyls (e.g., Sovol ($C_{12}H_5Cl_5$), Hexol (a mixture of 20% $C_{12}H_5Cl_{5n}$ and 80% C_4Cl_6), Sovtol (a solution of Sovol in trichlorobenzene), trichlorodiphenyl $(C_{12}H_7Cl)$, Pyranol, Chlorophen, Orophen, etc.). General structural formulas of the molecules of substituted hydrocarbons that form the basis of synthetic dielectric liquids are shown in Fig. 2.

The most prominent representatives of substituted hydrocarbons are polychlorinated biphenyls (PCBs). In 1930, it was established that PCBs possess high fire resistance and better dielectric properties than mineral oil. Until the 1960s, PCBs were used as dielectric liquids for transformers and other electrical equipment, such as capacitors. However, by 1970, environmental concerns were raised due to their high toxicity. Extensive research of PCB-based liquids has led to

Fig. 2. Chemical structures of the molecules of substituted hydrocarbons: *a*, chlorinated diphenyls; *b*, fluorinated hydrocarbons.

new claims about such liquids being extremely toxic organic pollutants of the environment. PCBs themselves, as well as their combustion products, have a highly negative impact on all living things, can accumulate in both plant and animal organisms, and are capable of causing autoimmune diseases not only in the current generation, but also be transmitted hereditarily to a new generation [7]. Therefore, due to high toxicity of PCBs, a declaration was adopted in 1978 prohibiting the use of these liquids and requiring all equipment operating with the use of PCBs to be discarded [7]. In Russia, the production of PCBs continued until 1993, after which it was also banned [7]. Currently, activities are underway to decommission and dispose of any PCB-containing equipment.

In the 1970s, *silicone fluids* were developed to replace toxic PCBs. Silicone oil is a silicon-based polymer, the structure of which consists of carbon, oxygen, and hydrogen at-

TABLE 2. Key Manufacturers of Mineral Dielectric Oils

Manufacturer	Trade name	Type of dielectric fluid	Application
Nynas AB (Sweden)	Nytro 10XN Nytro 11GX Nytro 11GBX Nytro 4000X Nytro 4000A Nytro Gemini X Nytro Lyra X Nytro Orion II Nytro Izar II Nytro Bear Nytro Libra Nytro Taurus	Naphthenic mineral oil	Power and measurement transformers, reactors, input devices $(\leq 1150 \text{ kV})$, switches
Ergon International (Belgium)	HyVolt	Same	Power transformers, switches
PetroChina Company Limited (China)	Kunlun	Same	Power transformers, reactors, switches
Royal Dutch Shell (Netherlands)	Shell Diala S4 ZX-1	Same	Capacitors, transformers, power cables
Calumet Specialty Products (USA)	CALTRANTM CALTRAN™ 60 - 00 Group U VOLTESSOTM	Same	Power transformers, reactors, switches
NK ROSNEFT (Russia)	Paraffinic mineral oil GK		Power and measurement transformers, reactors, input devices $(\leq 1150 \text{ kV})$, switches
LUKOIL (Russia)	VG	Same Power and measurement transformers, reactors, switches	
Total (France)	Total ISOVOLTINE II (non-inhibited) ISOVOLTINE II X (inhibited)	Naphthenic mineral oil	Power and measurement transformers, switches
REPSOL (Spain)	Repsol Electra 3X Repsol Tensiyn Centauro X		Power and distribution transformers, switches

Fig. 3. Chemical structure of the molecules of dimethylpolysiloxanes (2D structural formula).

oms (Fig. 3). Typically, polydimethylsiloxane chains include up to $40 - 50$ Si-O bonds.

Unlike PCBs, silicone oils do not exhibit toxicity. Silicone fluids possess dielectric characteristics similar to those of mineral dielectric oil and demonstrate good heat dissipation properties, high oxidation resistance, exceptional thermal stability, and reduced flammability [8].

The high cost of silicone oils, their high viscosity at freezing temperatures, and virtually zero biodegradability prompted a further search for an "ideal" dielectric liquid. Alongside silicone oils, research in the field of esters was conducted, resulting in synthesizing the first *synthetic ester* in 1977 and starting the mass production of such esters in 1985 [9].

From a chemical composition standpoint, synthetic esters mainly contain ester compounds of polyhydric alcohols (e.g., pentaerythritol and neopentyl glycol) with carboxylic acids. The most common esters are those based on tetrahydric alcohol (pentaerythritol), the structure of which is shown in Fig. 4. In this case, the number of carbon atoms in the carboxyl radical can range from C_5 to C_{22} , while the structure of these radicals can be either linear, or isomeric. Furthermore, all four ester bonds of pentaerythritol may contain the same fatty acids $(R_1 = R_2 = R_3 = R_4)$ or four different radicals $(R_1 \neq R_2 \neq R_3 \neq R_4)$, which means different fatty acids with different structures [10]. Synthetic esters that are manufactured today in accordance with the European standard IEC 61099, should not contain any halogen atoms.

Extensive research into the properties of synthetic esters has demonstrated that these fluids possess excellent dielectric characteristics, outstanding chemical stability, and a lower chilling point. At room temperature, synthetic esters absorb moisture 20 to 30 times more (800 to 1300 ppm) than mineral dielectric oil without a significant reduction in breakdown voltage. Compared to dielectric petroleum-based

Fig. 4. Chemical structure of the molecules of synthetic ester (pentaerythritol and carboxylic acids) [10] (2D structural formula).

oils, the ignition and flash points of synthetic esters are significantly higher. Unlike mineral oils, synthetic esters are much more susceptible to biodegradation (at least 80%). However, their cost is considerably higher than that of petroleum-based oils.

Currently, major producers of synthetic esters include M&I Materials Ltd Hibernia Way Trafford Park (UK), Cargill Incorporated (USA), and NYCO (France), which manufacture fluids under the trade names Midel 7131, Envirotemp® 360 fluid, and Nycodiel, respectively (Table 3).

The pursuit of creating a fully biodegradable insulating liquid began to materialize in the 1990s. *Plant-based oils* are considered the most promising and accessible biodegradable dielectric alternative to mineral oils. They are naturally extracted from the seeds, flowers, or fruits of various plants (e.g., castor, sunflower, rapeseed, olive, palm, flax, coconut, etc.). Plant-based oils are classified as environmentally friendly, renewable fluids. Numerous studies have established that plant-based oils are highly biodegradable (above 95%), less toxic, and have high ignition points (in excess of 300°C) [12]. Compared to mineral oils, plant-based oils dissolve a significant amount of water (about 500 ppm), while maintaining an acceptable level of breakdown voltage [13]. Plant-based oils can compensate for most of the major drawbacks associated with the use of petroleum-based oils, such as a high likelihood of ignition in the event of an accident and adverse environmental consequences in the case of oil leakage. However, the presence of a significant amount of

TABLE 3. Key Manufacturers of Synthetic Esters

Manufacturer	Trade name	Type of dielectric fluid	Application
M&I Materials Ltd Hibernia Way Trafford Park (UK)	Midel 7131	Synthetic biodegradable pentaerythritol-based ester	Power transformers up to 433 kV
Cargill Incorporated (USA)	Envirotemp [®] 360 fluid	Synthetic biodegradable ester	Unsealed traction and distribution transformers
Total (France)	Total ISOVOLTINE BIO	Same	Traction and distribution transformers, resistors, switches
Nyco Nycodiel 1244 NYCO (France) Nyco Nycodiel 1255 Nyco Nycodiel 1233		Same	Traction and distribution transformers, wind-powered generator transformers
REPSOL (Spain)	Repsol Bio Electra Synth	Same	Traction and distribution transformers

various unsaturated fatty acids makes plant-based oils vulnerable to rapid oxidation [14]. Consequently, the thermal instability of the oils increases with the transition from monoto tri-unsaturated acids. In other words, oil containing any tri-unsaturated fatty acid is more susceptible to oxidation than oil with an equivalent content of mono-unsaturated acid. The presence of metallic catalysts (e.g., copper, iron, zinc, etc.) significantly worsens the situation [12].

As a result of comprehensive research, it was concluded that dielectric liquids based on a variety of plant-based oils do not meet certain requirements for use as insulating and cooling media in power transformers. For instance, coconut oil cannot be used in cold conditions or at high voltages, as it crystallizes at freezing temperatures and rapidly oxidizes once exposed to atmospheric oxygen, temperature, and high voltage, which turns it into a thick, dark mass due to polymerization [13]. A similar situation occurs with other plantbased oils, particularly palm and rapeseed oils [13, 15].

Subsequently, a solution was found to improve the properties of plant-based oils, including an increase in their oxidation resistance, which had to do with esterification of fatty acids. It was found that the most thermally and chemically stable compounds are glyceryl fatty acid esters, which led to the creation of a new type of dielectric liquids — *natural esters*. The basis for the production of natural esters is primarily soybean, rapeseed, and sunflower oils [13, 16].

Chemically, natural esters belong to fats, which represent triglycerides — esters of a trifunctional alcohol — glycerol and three high molecular weight fatty acids (Fig. 5). Fatty acids typically have an unbranched carbon atom chain — radicals R_1 , R_2 , and R_3 . The length of these R_1 , R_2 , and R_3 chains can consist of several carbon atoms (typically, from C8 to C_{22}), which may contain from zero to three double bonds. The structure of a natural ester may contain identical $(R_1 = R_2 = R_3)$ or different $(R_1 \neq R_2 \neq R_3)$ acid residues.

The use of such "green" and environmentally friendly dielectric liquid in high-voltage transformers began at the end of the 20th century [13]. In 1996, the first transformer prototype was constructed, which utilized a natural ester as the dielectric medium. Mass production of the transformers filled with "green" dielectrics (natural esters) commenced in 1999. In the same year, the American division of the Swiss company ABB obtained the first patent for a commercial dielec-

Fig. 5. Chemical structure of the molecules of natural ester (2D structural formula).

tric liquid based on plant-based oil under the trade name BIOTEMP [16]. Shortly thereafter, Cargill Incorporated (USA) introduced a dielectric liquid based on plant-based oil under the trademark Envirotemp $FR₃$ [17]. In October 2019, the Swedish company Nynas AB expanded the range of commercial natural esters available on the market with a biological liquid hydrocarbon dielectric, manufactured from renewable resources, under the trade name NYTRO BIO 300X [18].

Currently, major producers of biodegradable natural esters and plant-based oils are international companies listed in Table 4.

Along with such positive properties as high biodegradability (above 95%), significant moisture absorption capacity, low toxicity, high ignition point (in excess of 300°C), and renewability, natural esters are less competitive than mineral dielectric oils in terms of cost, chill point, density, viscosity, and especially oxidation resistance [17]. Therefore, in order to improve the properties of dielectric liquids, including natural esters, nanotechnologies have been steadily integrated lately in the creation of an "ideal" dielectric liquid.

The concept of a "nanodielectric" liquid was proposed by a number of scientists as early as in 1994, however, this field has been actively developing since 2008 [19]. Nanofluids consist of a base liquid (e.g., mineral dielectric oil, synthetic ester, or natural ester) containing dispersed and suspended nanoparticles ranging in size from 1 to 100 nm $(1 \text{ nm} = 10^{-9} \text{ m})$. Nanosized additives are characterized by a

large interphase surface area, which prevents them from settling in the liquid medium, thereby transforming some characteristics of the base liquid. Nanoparticles of certain substances (at specific concentrations) are intended to improve the dielectric properties, thermal conductivity, as well as thermal and chemical stability of the dielectric liquid [20]. Table 5 provides some information about nanoparticle substances and their impact on the properties of the dielectric liquids.

Currently, extensive efforts are being made to create and study nanofluids, while a number of unresolved issues remain concerning their application in high-voltage equipment. Moreover, both positive and negative effects of the nanoparticles on the base dielectric liquid can be noted. For instance, according to the data analysis, the addition of nanoparticles (e.g., $Fe₃O₄$, TiO₂, Al₂O₃) reduces the resistivity of mineral dielectric oils and increases dielectric losses $[19 - 25]$. The viscosity of nanofluids is higher than that of any base dielectric liquid, which could complicate the startup of a transformer or any other equipment after extended downtime at freezing temperatures. No information is available about the compatibility of such nanofluids with the structural materials of transformers or other high-voltage equipment. No studies have been carried out regarding such important operational aspects as the mechanism of discharge in nano-modified oil, process of creeping discharge on the

surface of cardboard immersed in nanofluid, effect of nanoparticles on the development of streamers, etc.

Moreover, some studies have yielded contradictory results, apparently due to the varying concentrations of added nanoparticles. This indicates a lack of clear understanding of which nanoparticles (substance-wise) and in what concentration would result in a dielectric liquid with strictly defined characteristics. Consequently, at present, there is no standard for the creation, application, or quality assessment of either new or in-service nanofluids.

Nevertheless, on-going research expands our knowledge of the fundamental mechanisms of interaction between nanoparticles and various base liquids (e.g., mineral oils, synthetic and natural esters). The constantly increasing demand for electricity will lead to a demand for oil-filled transformers with higher performance characteristics. Nanofluids, possessing superior thermal and dielectric properties, can aid in the creation of more powerful transformers without increasing their size, which is an economically advantageous solution for manufacturers of electrical equipment. However, this is all in the future, as the research in the field of finding the optimal nanofluid formulation is still in the early stages.

Despite the trending increase in the production of power transformers with solid insulation, the overwhelming number of power transformers in Russia and other countries are currently utilizing an "oil-cellulose" insulation system. The advantage of using liquid insulation over gas or solid insula-

Nanoparticle	Concentration in base fluid	Base fluid	Observed effect
Titanium (IV)	$1 wt. \%$	Mineral oil	- Increase in breakdown voltage by a factor of 1.4.
oxide $(TiO2)$			- Deterioration of the dielectric properties of the mineral dielectric oil with a
			high content of TiO ₂ particles (at levels of $5 - 40\%$).
			- Reduction in oil resistivity.
			- Enhancement of oil thermal stability.
Mixture of Al_2O_3	0.01 wt.%	Same	- Enhancement of oil thermal conductivity.
and oleic acid (AlN)			— Slight increase in viscosity.
Aluminum oxide	20 mg/liter	Same	- Enhancement of the dielectric strength of the oil.
(Al_2O_3)			- Reduction in oil resistivity.
			- Increase in dielectric losses.
Silicon oxide $(SiO2)$	$1 wt. \%$	Same	- Enhancement of the dielectric strength of both fresh and oxidized dry oil.
			- Significant reduction in breakdown voltage at higher water content of the oil.
Iron(II, III) oxide $(Fe3O4)$	$1 wt. \%$	Mineral oil;	- Dielectric breakdown voltage of the modified oil increased by 1.26 times
(mixture of FeO and		mixture of natural	compared to unmodified oil.
$Fe2O3$, magnetite)		ester and	— More pronounced improvement in the dielectric properties of the mineral di-
		mineral oil	electric oil compared to a mixture of natural ester and oil.
			- Reduction in resistivity of the mineral oil.
			— Increase in the dielectric losses of the oil.
Zink oxide (ZnO)		Soybean and palm oil	- Enhancement of breakdown voltage of the modified plant-based oil.
Nano-diamond	$0.12 \text{ wt.} \%$	Mineral oil	- Enhancement of the dielectric strength and service life of the mineral dielec-
			tric oil.
			- Increase in thermal conductivity of the modified mineral oil by 14.5% com-
			pared to unmodified oil.
			— Slight increase in viscosity.
Fullerene (C_{60})	$150 - 250$	Same	- Maintaining a consistently high breakdown voltage during the aging process.
	mg/liter		- Reduction in dielectric losses during the aging process.
			— Decrease in the water absorption capacity of the oil at higher C_{60} content.

TABLE 5. Effect of Nanoparticles of Certain Substances on the Characteristics of Dielectric Fluids [19 – 25]

tion consists in more effective heat removal from the heated parts of the transformer. In addition, dielectric liquids perform few more key functions in power transformers, which include the following: insulating current-conducting parts; protecting solid insulation from moisture; quenching spark and arc discharges; serving as acoustic damping and diagnostic media.

CONCLUSIONS

1. Globally, the majority of power transformers are filled with transformer oil. Gradually, transformer oil is being replaced by other liquid dielectrics, especially in transformers rated up to 110 (150) kV. Along with other important factors, the reliable and uninterrupted operation of oil-filled power transformers is ensured by good dielectric properties of the insulation system, which includes both solid and liquid insulation.

2. To ensure successful operation of oil-filled transformers, modern power engineering imposes a number of requirements on the properties of liquid dielectrics. Thus, the dielectric liquid must possess high dielectric strength, good thermal conductivity, adequate chemical and thermal stability, minimal toxicity, high biodegradability, proper viscosity, and high fire-resistant properties.

3. Transformer oils, which have been used quite successfully in oil-filled equipment for over 130 years, have several disadvantages: they are produced from a non-renewable natural resource (petroleum); have lower chemical stability compared to synthetic fluids; have low fire-resistant properties and low biodegradation ability.

4. During a period from 1930 to 1970 (in the USSR and CIS — until 1993), synthetic fluids, such as polychlorinated biphenyls (PCBs), were used as non-flammable dielectric liquids in distribution power transformers and capacitors. However, due to their high toxicity and negative impact on the environment, their use was banned in 1978 (in Russia in 1993).

5. Currently, to ensure the reliable operation of power transformers while maintaining an environmentally friendly approach, dielectric biofluids are being increasingly considered, which offer several advantages over petroleum-based mineral dielectric oil. Synthetic ester-based fluids exhibit enhanced fire-resistant properties, have significantly lower toxicity, high degree of biodegradability, high chemical stability, high water solubility, and dielectric strength. They are primarily used in transformers installed in locations having stricter requirements for fire safety and environmental protection (e.g., densely populated areas, tunnels, subways, oil platforms, etc.).

6. Natural ester-based fluids have dielectric properties similar to those of synthetic esters, but are inferior to mineral oils and synthetic esters in terms of chemical stability. Therefore, their use is currently limited to sealed high-voltage transformers.

7. Since 2008, researchers have been actively creating and studying dielectric properties of nanofluids (base fluids include oil and esters with added particles of certain substances measuring several nanometers in size). However, the dielectric properties of nanofluids are not yet fully understood, and their behavior in operating environments (under the exposure to high voltages, elevated temperatures, etc.) is not completely clear.

8. Despite the clear advantages of alternative dielectrics (e.g., natural and synthetic esters, nanofluids), there are several main factors restraining their widespread application in oil-filled power transformers, such as higher cost compared to transformer oil, lack of comprehensive information regarding the change in dielectric properties, and absence of regulatory documents detailing maintenance and diagnostics of their condition during operation.

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