

Dissolution of Bubbles with Diagnostic Gases in Insulating Liquids

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Abstract — Bubble dissolution in rapeseed, castor, mineral transformer and silicone oils is recorded optically. Diffusion coefficients of diagnostic gases (hydrogen, methane, ethane and ethylene) are determined by comparison of experimental data of bubble dissolution with computer simulation. Differences with previous data are in more careful preparation of clean oils and gases that leads to higher values of coefficients.

Keywords — transformer oil, hydrogen, diagnostic gases, dissolution, viscosity, diffusion coefficient.

I. INTRODUCTION

Study of bubble dissolution is important for liquid electrical insulation. At present the most commonly used method for condition assessment of high voltage electrical equipment with paper-oil insulation is chromatographic analysis of the gases dissolved in the oil (DGA). The analysis procedure can be divided into three stages: sampling of electrical insulating liquid, sample transport, extracting gas from the liquid and analysis performance. During the whole analyzing period the main task is to provide sample representativeness of electrical insulating liquid, especially equality between gas concentrations in composition which are in the tank to gas concentrations inside the sampler at the moment of probe injection into chromatograph. Gas diffusion in electrical insulating liquid directly influences on the representativeness of the sample at all stages of diagnostics. Determination of the diffusion coefficients of diagnostic gases in electrical insulating liquids is the task of great importance and significance.

Coefficients of diagnostic gas diffusion in transformer oil were determined in paper [1] by registration of pure gas bubbles dissolution in degassed liquid. However, nowadays several different liquids are used in electrical engineering. E.G. ABB corporation has a department producing experimental transformer equipment using best oils in terms of fire safety and ecology - vegetable oils [2].

The proposed research aim is determination of the diffusion coefficients for the main diagnostic gases dissolved in several types of oils.

II. MODEL

A. Idea of measurement

The computed curve of bubble dissolution should compare with experimentally registered one. Firstly the equation of

diffusion is solving with approximately appropriate diffusion coefficient. Initial bubble size is chosen equals to experimental measured one for each case of bubbles, gases and kind of liquid. If experimental and computed curves for each case differ one from other more the 5 % then diffusion coefficient in next computation is changed. After two or three steps of coefficient fitting the curves are practically the same and diffusion coefficient of this computation step is took into consideration as required one.

B. Mathematical model

The nonstationary equation of diffusion in a spherically symmetric medium (Fig.1) appears as follows:

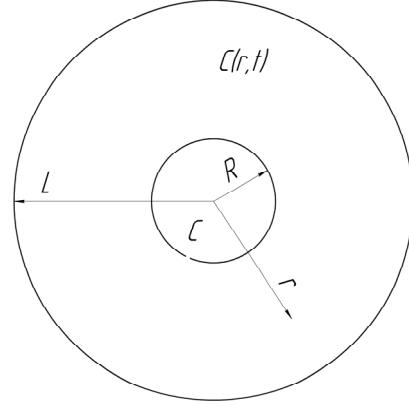


Fig. 1. Graphical representation of model. L - remote boundary, R – current bubble radius, $C(r,t)$ – gas concentration in liquid on point r and the time t, C – gas concentration in bubble.

$$\frac{d}{dt} C(r,t) = \operatorname{div}(D \cdot \operatorname{grad}(C(r,t))) \quad (1)$$

where concentration c depends on current radius r and time t . Since, according to the condition of the problem, the bubble at any moment differs only in volume, there is no need to include it in the calculation region. In this case, the problem should be solved like a boundary value problem with a mobile boundary (bubble) with the corresponding boundary conditions:

$$\left. \frac{\partial}{\partial n} C(r,t) \right|_{r=L} = 0 \quad (2)$$

$$C(r,t) \Big|_{r=R(t)} = k \cdot C \quad (3)$$

where L is a sufficiently remote boundary, $R(t)$ is the bubble radius, k is the solubility coefficient, and C is the gas concentration in a bubble. It is obvious that in the course of diffusion, the bubble volume decreases and the liquid surrounding a bubble moves behind the bubble boundary.

Hence, the derivative $\frac{d}{dt} C(r,t)$ in (1) should be understood as substantial. As well, it is obvious that the following integrobalance relations need to be fulfilled:

$$C \cdot V(t) + 4\pi \int_{R(t)}^L C(r,t) r^2 dr = C \cdot V_0 \quad (4)$$

expressing the law of conservation of mass at a time. In (4),

$$V(t) = \frac{4\pi}{3} R^3(t) \text{ and } V_0 = \frac{4\pi}{3} R^3(t_0)$$

are the current and initial bubble volumes, respectively. To solve Eq. (1) with boundary conditions (2), (3), we use the finite element method.

These basic equations were obtained for the case of absolutely clean bubble and absolutely clean liquid. The harmful impurity either for liquid or bubble is air. Some cases concerning air influence were treated by computation or experimentally. Air concentration c_a in liquid should leads to air diffusion into bubble and slow down process of bubble dissolution at the last stages of dissolution, but air concentration in bubble should leads to immediately deceleration of dissolution.

III. EXPERIMENTAL METHOD, RESULTS

Both experimental data and mathematical model are used for determination of the diffusion coefficients. The bubble of pure diagnostic gas is injected into degassed oil. The experimental test cell is shown in Fig. 2. The process of bubble dissolving in the oil is recorded. Then the video picture is splitted into frames in specified time periods. After that the bubbles size is determined by means of graphics editor. The data are used to build curves showing the diagnostic gas dissolution.

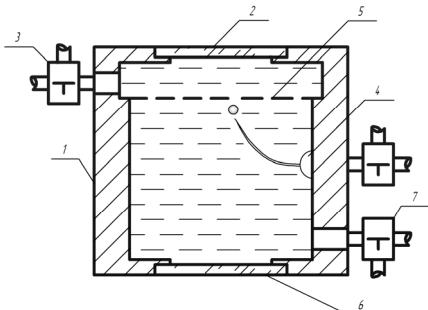


Fig. 2. Test cell. Vertical cylinder (1), glass cover (2) and bottom (6), mounted into the wall needle (4), valves (3) and (7), the mesh (5) which enables to keep the gas bubble within the fluid. Under bottom is light source; above cover is microscope, which eyepiece has video camera.

Typical frames taken from video film of bubble dissolving on the mesh is shown in Fig. 3.

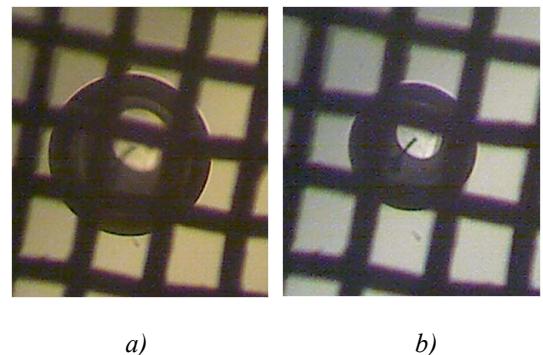


Fig.3. Initial bubble on the mesh (a), bubble on the finish stage of dissolution on the mesh.

B. Rapeseed oil

Typical experimental data for bubble dissolving in rapeseed oil presented in Fig 4.

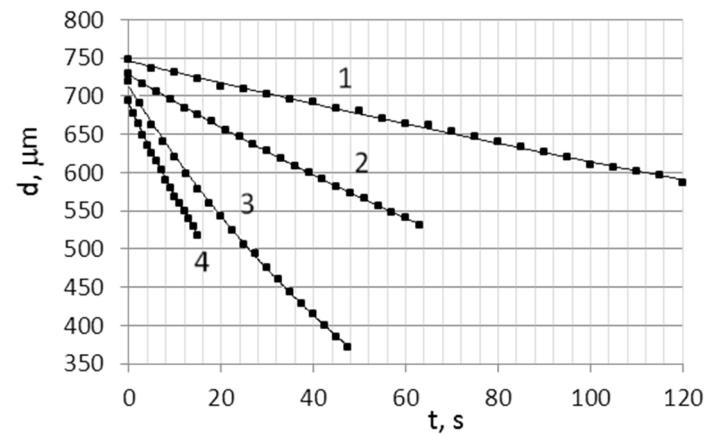


Fig. 4. Experimental curves representing dissolution of gas in rapeseed oil: 1 – hydrogen; 2 – methane; 3 – ethane; 4 – ethylene.

One could see that ethane is most fast dissolving gas, next gas is ethylene, then methane and hydrogen. More than 10 experimental curves for each gas were mathematically processed to fit experimental and computed curves.

Fitted gas diffusion coefficients for rapeseed oil are presented in Table I. Solubility coefficients were taken from [7].

TABLE I. COEFFICIENTS OF GAS DIFFUSION IN RAPESEED OIL

	Average value of diffusion coefficient, m ² /s			
	Hydrogen	Methane	Ethane	Ethylene
Rapeseed oil	10^{-8}	3×10^{-9}	1.3×10^{-9}	1.2×10^{-9}

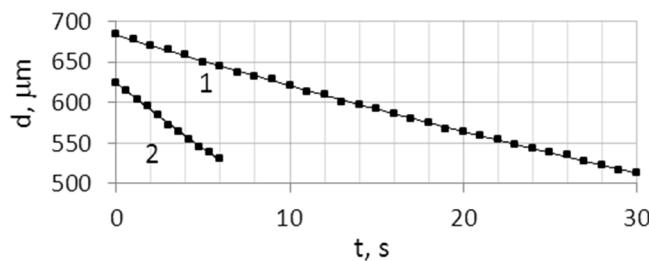
C. Silicone oil

Fig.5. Experimental curves of gas dissolution of in silicone oil: 1 – hydrogen; 2 – methane.

In silicone oil methane show more faster dissolution in comparison with hydrogen (Fig.5). Fitted diffusion coefficients are shown in Table III.

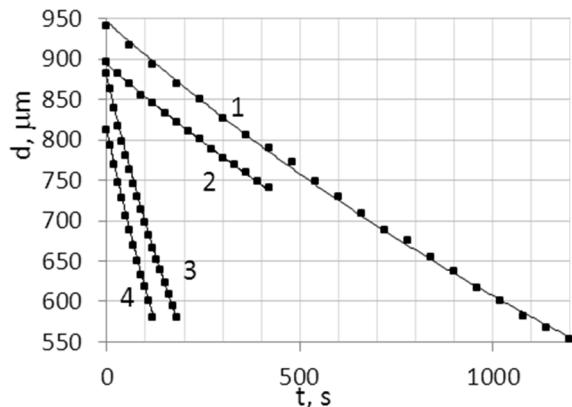
D. Castor oil

Fig.6 Experimental curves of gas dissolution of castor oil: 1 – hydrogen; 2 – methane; 3 – ethylene; 4 – ethane

In castor oil gases shows the same succession as gases in rapeseed oil (Fig.6). Fitted diffusion coefficient one could to obtain for hydrogen only (see Table IV), because solubility coefficient for castor oil was measured for this gas only [8].

IV. DISCUSSION

All data presented in fig. 4–6 shows that hydrogen is slowest gas in all liquids. What was the cause for this behavior? Molecule of hydrogen has least radius in comparison with other molecules. That is why it seems that rate of dissolution should have highest value. But in this case other characteristic plays more important role, namely low solubility of hydrogen. Table II, taken from [7], shows that hydrogen has from one to two orders of magnitude lesser solubility than other diagnostic gases.

TABLE II. COMPARISON OF GAS SOLUBILITY IN TRANSFORMER OIL AND RAPSEED OIL AT TEMPERATURE 20 °C

Gas name	Solubility in transformer oil	Solubility in rapeseed oil
H ₂	0,05	0,06
CO	0,12	0,12
CH ₄	0,40	0,34
C ₂ H ₄	1,75	1,58
C ₂ H ₆	2,78	1,85
C ₂ H ₂	1,20	2,89
CO ₂	1,08	1,36

One could see that solubility coefficient has stronger effect on bubble dissolution rate than size of molecule.

The authors noticed the coefficients mismatch between obtained gas diffusion coefficients in rapeseed oil and gas diffusion coefficients determined earlier for transformer oil [1] (Table III).

TABLE III. COMPARISON OF PRESENT EXPERIMENTAL DATA WITH PUBLISHED EARLIER

	Average value of diffusion coefficient, m ² /s			
	Hydrogen	Methane	Ethane	Ethylene
Rapeseed oil	10 ⁻⁸	3 x 10 ⁻⁹	1.3 x 10 ⁻⁹	1.2 x 10 ⁻⁹
Transformer oil [1]	3.3 x 10 ⁻⁹	10 ⁻⁹	0.4 x 10 ⁻⁹	-

Usually diffusion and viscosity have practically the same nature: stochastic movement of molecules. In case of diffusion it is random oscillations of guest and host molecules and in case of viscosity - random oscillations of host molecules only. That is why in case of rapeseed oil the diffusion coefficients should be less than in case of mineral transformer oil due to the well-known fact that rapeseed oil viscosity is several times greater than transformer oil viscosity. To explain this mismatching the decision to investigate transformer oil again was made. The following dissolution curves for hydrogen in transformer oil and rapeseed oil were obtained at the same procedure of bubble and liquid preparation (Fig 7).

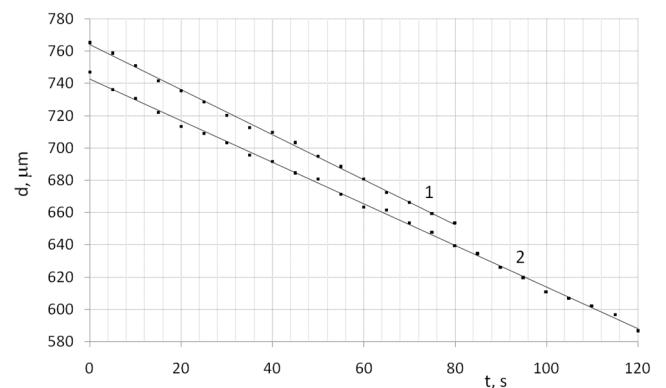


Fig. 7. Hydrogen dissolution curves: 1 – in transformer oil; 2 – in rapeseed oil.

The diagram shows that curves representing hydrogen bubble dissolution in transformer oil has steeper angle of

inclination. It means that hydrogen coefficient of diffusion in transformer oil is really greater than in rapeseed oil.

Some hypotheses were tested to clear up mismatching of data received. The most probable cause of mismatching is a little amount of dissolved air in case of insufficient liquid degassing during the previous experiment [5]. The basement of this assumption is in the fact that in present experiments more efficient vacuum system for degassing was applied. Mathematical model of bubble dissolution in oil containing dissolved air was developed. It showed that dissolved air effects on the dissolution rate at last dissolution stages when the bubble size is smaller than in Fig.3-7. However, when assumed that residual air is in not into liquid but in the bubble then gas-air bubbles dissolution must be slower even at initial stages of dissolution.

In the process of analysis of possibility for air to be inside the gas transmission tract it was found that possible cause was slow blowing of transmission path by pure gas. Needle used as gas transmission path was blown insufficiently. The reason was in great hydrogen diffusion in the air which resulted in hydrogen mixing with air at low blowing speed. Not pure gas came to degassed oil but a mixture of the gas and air. Diffusion coefficients for nitrogen and oxygen are lower. Therefore it dissolves slower retarding gas bubble dissolution. This assumption was proved experimentally, at different blowing speed it is possible to have different dissolution rate (Fig.8).

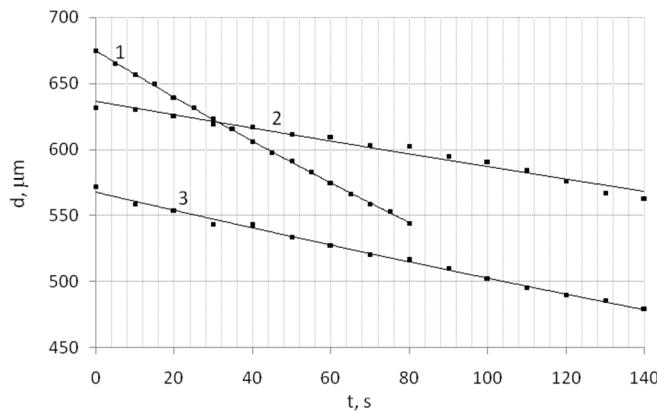


Fig. 8 Hydrogen dissolution curves in transformer oil: 1 –repeated blowing of gas supply channel; 2 – single blowing; 3 –result of [1].

To calculate diffusion coefficients of basic diagnostic gases in rapeseed and transformer oils the dissolution coefficients of basic diagnostic gases [3] were used. For castor oil dissolution coefficients were taken from [4]. Data for silicone oil were obtained in our laboratory [5]. All calculated diffusion coefficients are presented in Table IV.

TABLE IV. DIFFUSION COEFFICIENTS

	Average value of diffusion coefficient, m ² /s			
	Hydrogen	Methane	Ethane	Ethylene
Rapeseed oil	10^{-8}	3×10^{-9}	1.3×10^{-9}	1.2×10^{-9}
Transformer oil	1.4×10^{-8}	2.3×10^{-9}	1.1×10^{-9}	1.2×10^{-9}
Castor oil	7.5×10^{-9}			
Silicon oil	2.3×10^{-8}	9×10^{-9}		

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