

Dissolution of Diagnostic Gas Bubbles in Transformer Oil

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Abstract—This work is devoted to the problem of studying the propagation processes of the main diagnostic gases in oil-filled electric equipment. By experimentally and numerically simulating bubble dissolution, the diffusion coefficients of three gases have been determined: hydrogen, methane, and ethane in transformer oil. The value of the diffusion coefficient of hydrogen, 3.3×10^{-9} m²/s is not abnormally large, which has important practical value for diagnostics of oil-filled electric equipment.

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INTRODUCTION

Chromatographic analysis of gases dissolved in oil (DGA) is considered the most informative method of diagnosing high-voltage oil-filled electric equipment. In this, an oil sample is taken from the working equipment; gases (hydrogen, methane, ethane, ethylene, and acetylene) are extracted from it. According to the concentration values, the rate of their increase, and, in particular, the concentration ratios, the working capacity of the device is determined. Usually, in transformers, the place from which samples are taken is located sufficiently far from places with high field strength values, where gas formation processes occur. The reliability of the obtained concentration values depends on how uniformly the dissolved gas is distributed and to what extent the gas concentrations correspond to each other in the sampling and generation regions. In turn, gases are distributed over the oil volume owing to diffusion and the appearance of flows. In addition, diffusion plays an important role in preserving gases in triers, as well as their extraction for analysis. The most informative and problematic gas during analysis is hydrogen, since, on the one hand, it forms under nearly any energy processes occurring in electric equipment, and on the other hand, due to the small size of the hydrogen atom, it is intuitively supposed that it easily “departs” from the oil sample at all diagnostic stages.

However the diffusion coefficient of hydrogen in transformer oil has been established, according to the authors, insufficiently correctly. The diffusion coefficient of hydrogen in oil was determined only in [1], and their obtained value of 10^{-7} m²/s is improbably high. For comparison, we show a value of the self-diffusion coefficient of water in water of 2.5×10^{-9} m²/s [2]. In oil, if we consider its increased viscosity, the diffusion coefficient of molecules of comparable size should be even less. Even if the size of a water molecule is slightly more than the size of a hydrogen molecule,

all the same, the abnormally large value of the diffusion coefficient of hydrogen in oil seems improbable.

The diffusion coefficient of other diagnostic gases have not been determined at all.

The goal of this work is to determine the diffusion coefficient of hydrogen, methane, and ethane in transformer oil.

TECHNIQUE OF APPROXIMATE ESTIMATION

Earlier [3], dissolution of gas bubbles in a liquid was considered and the expression describing the time dependence of the radius of a solitary motionless bubble in an infinitely large volume of a liquid unsaturated by a gas was obtained:

$$\frac{dR}{dt} = kD \left(\frac{1}{R} + \frac{1}{(\pi D(t-t_0))^{1/2}} \right). \quad (1)$$

Here R is the current value of the radius of a dissolved bubble; D is the diffusion coefficient of a gas in a liquid; $t - t_0$ is the time that elapsed after the onset of dissolution; k is the gas-solubility coefficient (the ratio of the gas concentration in a liquid c to the concentration in the gas phase under conditions of equilibrium between phases). This expression for the case of small bubbles (and large diffusion coefficients) becomes simpler due to neglect of the second term in brackets, which after simple transformations allows us to obtain a simple expression to determine the diffusion coefficient:

$$D = \frac{R^2(t_0) - R^2(t)}{2k(t-t_0)}. \quad (2)$$

To obtain data, it is necessary to know gas solubility k and to record the radii of a microbubble $R(t_0)$, $R(t)$ at the initial time moment and after a certain time $t - t_0$ has elapsed. This method was suggested and used for the first time in [4], where in such a way the diffusion coefficients of gases in water were determined.

However, in the general case, it is not always possible to neglect the nonstationary term in expression (1). As well, to measure the diffusion coefficients of gases in transformer oil, it is necessary to solve a quadratic equation relative to D in which it is necessary to substitute the measured values of time, radius, and the radius derivative over time.

MATHEMATICAL SIMULATION OF THE DISSOLUTION PROCESS

The nonstationary equation of diffusion in a spherically symmetric medium appears as follows:

$$\frac{d}{dt}c(r, t) = \text{div}(D \text{grad}(c(r, t))), \quad (3)$$

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:

$$\left. \frac{\partial c}{\partial n} c(r, t) \right|_{r=L} = 0, \quad (4)$$

$$c(r, t)|_{r=R(t)} = kC, \quad (5)$$

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 (3) should be understood as substantial. As well, it is obvious that the following integrobalance relations need to be fulfilled:

$$CV(t) + 4\pi \int_{R(t)}^L c(r, t) r^2 dr = CV^0, \quad (6)$$

expressing the law of conservation of mass at any moment. In (6), $V(t) = \frac{4\pi}{3} R^3(t)$ and $V^0 = \frac{4\pi}{3} R^3(t^0)$ are the current and initial bubble volumes, respectively.

To solve Eq. (3) with boundary conditions (4), (5), we use the finite element method [5].

An equivalent variational statement in Galerkin form for Eq. (3) with boundary conditions (4), (5) appears as follows:

$$\int_{R(t)}^L D \cdot \frac{\partial c(r, t)}{\partial r} \cdot \frac{d}{dr} \psi \cdot r^2 dr + \int_{R(t)}^L \frac{\partial c(r, t)}{\partial t} \cdot \psi \cdot r^2 dr = 0, \quad (7)$$

where $\psi = \psi(r)$ is the trial function.

We split the calculated region into finite elements (segments) $\Omega_i = [r_i, r_{i+1}]$. After discretization of Eq. (3) over time (for this purpose, we use a two-layer implicit

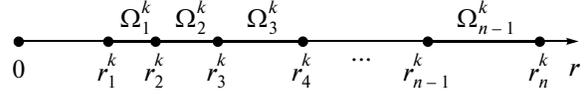


Fig. 1. View of calculated region.

scheme) on some k th time layer ($t = t^k$), the calculated area will have the form shown in Fig. 1, where $L = r_n^k$, $R(t^k) = r_1^k$. We connect to each node the basis function ψ_i^k , determined as

$$\psi_1^k(r) = \begin{cases} \frac{r_2^k - r}{r_2^k - r_1^k}, & r_1^k \leq r \leq r_2^k \\ 0, & r < r_1^k, r > r_2^k \end{cases},$$

$$\psi_n^k(r) = \begin{cases} \frac{r - r_{n-1}^k}{r_n^k - r_{n-1}^k}, & r_{n-1}^k \leq r \leq r_n^k \\ 0, & r < r_{n-1}^k, r > r_n^k \end{cases},$$

$$\psi_i^k(r) = \begin{cases} \frac{r - r_{i-1}^k}{r_i^k - r_{i-1}^k}, & r_{i-1}^k \leq r \leq r_i^k \\ \frac{r_{i+1}^k - r}{r_{i+1}^k - r_i^k}, & r_i^k \leq r \leq r_{i+1}^k \\ 0, & r < r_{i-1}^k, r > r_{i+1}^k \end{cases},$$

for $i = 2, \dots, n - 1$.

Then, the solution $c^k(r) = c(r, t^k)$ can be represented as $c^k(r) = \sum_{i=1}^n q_i^k \psi_i^k(r)$. Assuming $\frac{\partial c}{\partial t} \Big|_{t=t^k} \approx \frac{c^k(r) - c^{k-1}(r)}{t^k - t^{k-1}}$, $c^{k-1}(r) \approx \sum_{i=1}^n q_i^{k-1} \psi_i^k(r)$, choosing functions ψ as functions ψ_i^k , and substituting in (7) the expressions for $c^k(r)$ and $\frac{\partial c}{\partial t} \Big|_{t=t^k}$, without allowance for boundary condition (5), after elementary transformations, we obtain the matrix equation

$$\left(\mathbf{S}^k + \frac{1}{\Delta t^k} \mathbf{M}^k \right) \mathbf{q} = -\frac{1}{\Delta t^k} \mathbf{M}^k \mathbf{q}^{k-1},$$

in which $\Delta t^k = t^k - t^{k-1}$, and global matrices \mathbf{S}^k and \mathbf{M}^k are made up of local matrices of individual elements Ω_i^k of the type (for convenience, without the step index over time k)

$$S_i = \frac{r_{i+1}^2 + r_{i+1}r_i + r_i^2}{3\Delta r} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix},$$

$$M_i = \frac{\Delta r}{60} \begin{pmatrix} 2r_{i+1}^2 + 6r_{i+1}r_i + 12r_i^2 & 3r_{i+1}^2 + 4r_{i+1}r_i + 3r_i^2 \\ 3r_{i+1}^2 + 4r_{i+1}r_i + 3r_i^2 & 12r_{i+1}^2 + 6r_{i+1}r_i + 2r_i^2 \end{pmatrix},$$

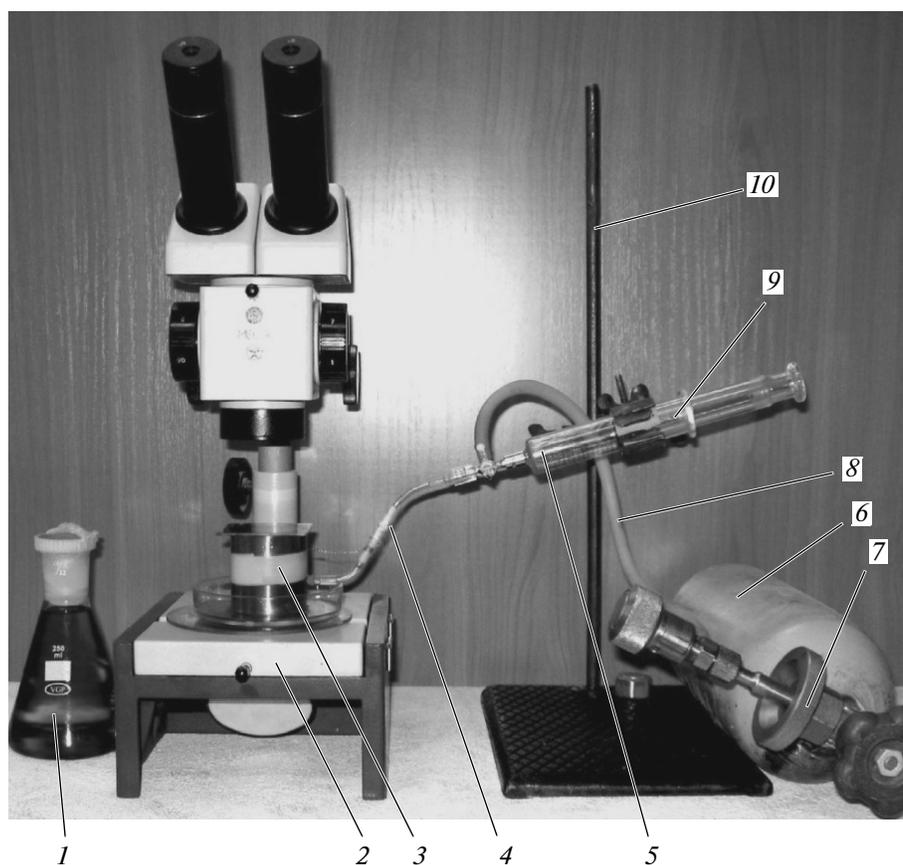


Fig. 2. Experimental setup: 1, conical flask; 2, object table of MBS-12 microscope; 3, experimental cell; 4, gas-intake tube (into the cell); 5 Elchrom sampler with individual grinding piston; 6, vessel with diagnostic gas; 7, fine-adjustment valve; 8, connecting tube; 9, sampler piston; 10, laboratory stand.

where $\Delta r = r_{i+1} - r_i$. To take boundary condition (3) into account, it suffices to fix the weight q_1^k and in a corresponding manner change the matrix of the system of linear algebraic equations (SLAE).

After finding weights q_i^k (SLAE solutions), requiring that relation (6) be satisfied, which after discretization takes the form

$$CV(t^k) + 4\pi \sum_{i=1}^{n-1} \int_{r_i^k}^{r_{i+1}^k} (q_i^k \psi_i^k + q_{i+1}^k \psi_{i+1}^k) r^2 dr = CV^0,$$

we find $V^k = V(t^k)$ and thereby the new boundary of the bubble $R^k = r_1^k = \sqrt[3]{\frac{3}{4\pi} V^k}$. Coordinates r_i^k , $i = 2 \dots n-1$, change according to the formula

$$r_i^k = r_i^{k-1} - (R^{k-1} - R^k) \frac{(R^{k-1})^2}{(r_i^{k-1})^2},$$

which corresponds to motion of the liquid surrounding the bubble [6].

This procedure, including assembling and solving the SLAE, as well as changing the grid, is performed before full stabilization of the boundary of the bubble R^k , after which there is a transition to the next time layer.

In the mathematical model, the value determined according to (1) is used as reference value D . Then the calculated graphs are compared with the graphs constructed from experimental data, and the value of the diffusion coefficient is refined. If after calculation, the graphs satisfactorily coincide with the specified value D , it is taken as the measured value of the diffusion coefficient.

EXPERIMENTAL SETUP

To conduct measurements, an experimental setup was developed whose general view is shown in Fig. 2. The cell of the experimental setup is a vertical cylinder (Figs. 2, 3) with a needle built into the wall to supply the diagnosed gas. The bottom and lid of the cell are made of glass, which makes it possible to record the dissolution dynamics of a gas microbubble. The volume is filled with decontaminated transformer oil and closed with the lid, which prevents penetration of air

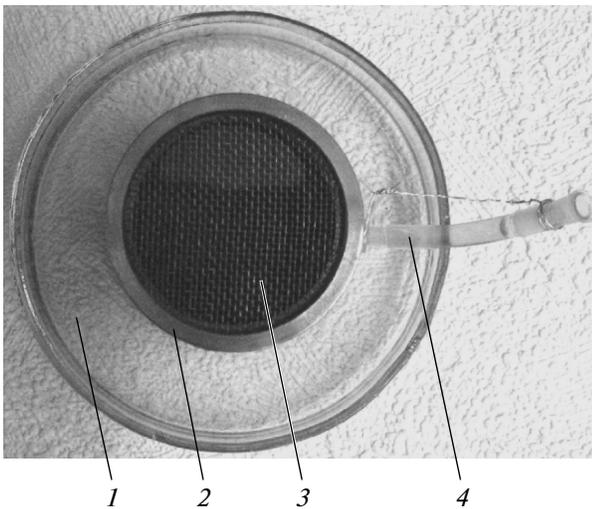


Fig. 3. Cell, view from above: 1, Petri dish; 2, body of experimental cell; 3, metal grid; 4, gas-intake tube.

into the system. The cell with the oil is placed under a microscope; via a gas-intake tube into the bulk of the oil, a bubble of the diagnosed gas is introduced through the needle, where it is suspended on the grid. The grid is a necessary element in system, because it makes it possible to consider a dissolved bubble motionless and spherical. The dissolution dynamics of a hydrogen microbubble is recorded with a videocamera equipped with microscope eyepieces instead of one eyepiece and it is connected to a personal computer.

Sampling and gas forcing is performed with an Elkhrom sampler [7], which represents a combination of a special all-glass syringe with an individual-grinding piston and vacuum T-valve. One exit is connected to the cell needle, and the second, to the cannister with the diagnosed gas. In order to avoid sharp jumps in pressure in the system, a fine-adjustment valve has been placed on the cannister. The syringe piston is greased with vacuum greasing to eliminate gas leak and entry of air into the system. Immediately prior to the experiment, the gas path was blown by the diagnostic gas to get rid of air in the hollows of tubes.

After recording of gas microbubble dissolution, the video is displayed by frame (Fig. 4) with a certain time interval. Breakdown into frames makes it possible to conduct necessary measurements and construct the time dependence of the radius and the radius derivative (Fig. 4).

The scale was determined by gauging the bubble diameters and grid wire (the wire diameter was 0.11 ± 0.01 mm). Figure 4 shows typical photographs of bubbles for two time moments. In data processing, the following values of solubility coefficients were used: for hydrogen, 0.049; for methane, 0.409; for ethane, 2.925. All data are taken from [8].

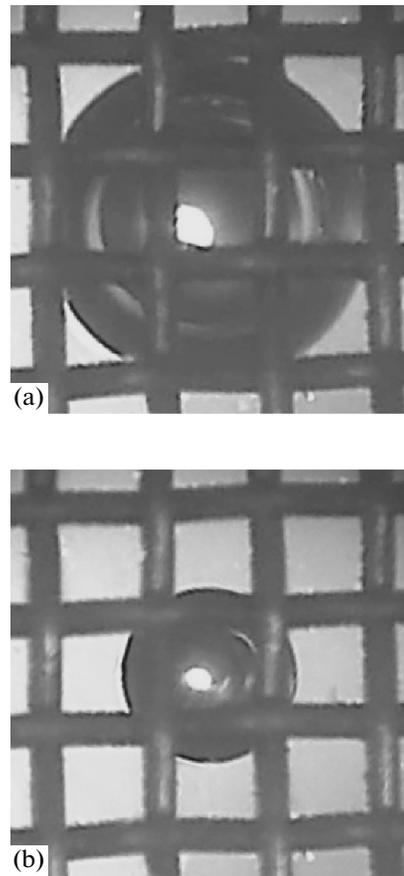


Fig. 4. Hydrogen bubble at the initial (a) and final (b) moments of recording.

RESULTS

Data processing using expressions (1), (2) for the series of measurements has made it possible estimate the diffusion coefficient in a first approximation. Figure 5 shows the results of calculating the diffusion coefficient of hydrogen in the case of dissolution of bubbles of various sizes. It can be seen that for small bubbles, expression (2) gives underestimated values, and for large bubbles, overestimated, in comparison to the values obtained with allowance for nonstationarity. As well, in the case of a simpler formula, the tendency toward an increase in the calculated value of the diffusion coefficient with increasing bubble radius is observed. This indicates the inapplicability of simplified formula (2) for calculation. Further, for numerical calculations the values obtained using expression (1) were used as the initial approximation.

Figures 6 and 8 show the experimental data, as well as the results of numerical calculation of hydrogen, methane, and ethane bubble dissolution.

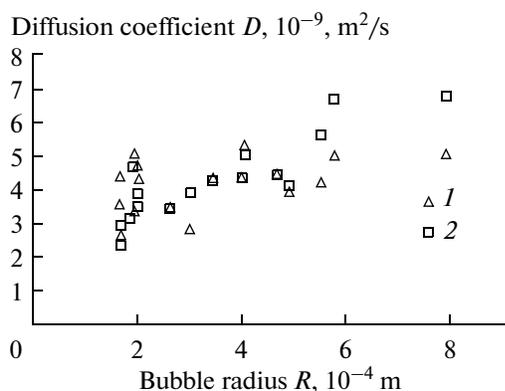


Fig. 5. Calculation of diffusion coefficient of hydrogen by simplified expressions: 1, expression (1); 2, expression (2).

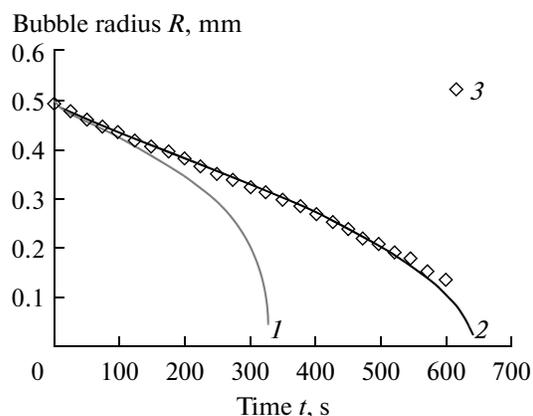


Fig. 6. Hydrogen bubble dissolution dynamics: 1, calculation without allowance for motion of liquid; 2, with allowance; 3, experimental points. The diffusion coefficient in numerical calculation $D = 3.3 \times 10^{-9} \text{ m}^2/\text{s}$; initial approximation $D = 3.9 \times 10^{-9} \text{ m}^2/\text{s}$.

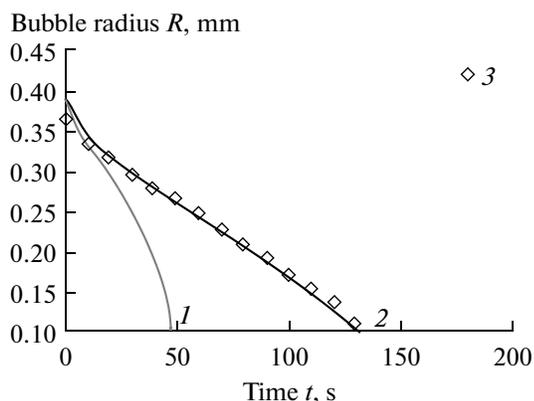


Fig. 7. Methane bubble dissolution dynamics: notations are the same as in Fig. 6. The diffusion coefficient in numerical calculation $D = 8 \times 10^{-10} \text{ m}^2/\text{s}$; initial approximation $D = 1.6 \times 10^{-9} \text{ m}^2/\text{s}$.

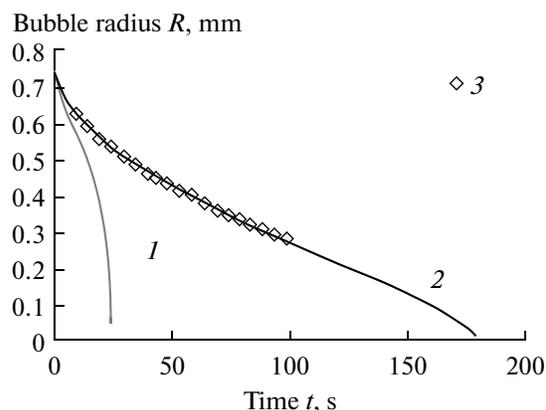


Fig. 8. Ethane bubble dissolution dynamics: notations are the same as in Fig. 6. The diffusion coefficient in numerical calculation $D = 4 \times 10^{-10} \text{ m}^2/\text{s}$; initial approximation $D = 1.9 \times 10^{-9} \text{ m}^2/\text{s}$.

DISCUSSION

From the obtained data, it follows that use of simplified expression (2) to calculate the diffusion coefficients is too rough and can only be applied for an estimate, mainly in the case of small bubbles. Expression (1) has also been obtained without motion of the liquid, and it gives plausible estimates when considering dissolution of hydrogen bubbles, but in the dissolution of other gases, the error is considerable. This is related to the small solubility coefficient of hydrogen and the large solubility coefficients of other gases. Therefore, motion of the liquid is insignificant in the case of hydrogen and significant in the case of methane and, in particular, ethane.

Indeed, the bubble dissolution curves obtained in computer simulation of dissolution with and without allowance for motion of the liquid are close for hydro-

gen (at least at the initial dissolution stage) and differ sharply for methane and ethane.

On the basis of processing the series of experiments, the following values of diffusion coefficients for the three main diagnostic gases in GK transformer oil have been obtained at room temperature:

for hydrogen, $D = (3.3 \pm 1) \times 10^{-9} \text{ m}^2/\text{s}$;

for methane, $D = (1 \pm 0.3) \times 10^{-9} \text{ m}^2/\text{s}$;

for ethane, $D = (4 \pm 1) \times 10^{-10} \text{ m}^2/\text{s}$.

We emphasize that the obtained values for the diffusion coefficient of hydrogen is almost two orders less than the diffusion coefficient determined earlier [1]. This is important for diagnostics of oil-filled equipment, since it makes it possible to confirm that hydrogen is not as volatile as was explicitly and implicitly supposed in chromatographic analysis of liquid elec-

tric-insulation decomposition products under the influence of operational factors.

To evaluate the behavior of the diffusion coefficients with changing temperature, a few experiments were performed at an increased temperature of up to 50°C. The diffusion coefficient increased approximately fivefold, which does not contradict the approximate Li–Chang expression [9]

$$D = \frac{1.84 \times 10^{-3}}{\eta r},$$

where r is the radius of a molecule of the dissolved substance. According to this estimate, the diffusion coefficient is related to viscosity by an inversely proportional dependence. Since oil viscosity drops approximately fivefold with a change in temperature from 20 to 50°C, this leads to a fivefold increase in the diffusion coefficient.

CONCLUSIONS

From the obtained data on the gas bubble dissolution dynamics, it is possible to conclude that the diffusion coefficient of hydrogen in transformer oil is not abnormally high either in value or in comparison to other gases. The values of diffusion coefficients of the main diagnostic gases in GK transformer oil have been obtained at room temperature: hydrogen $D = (3.3 \pm 1) \times 10^{-9}$ m²/s; methane $D = (1 \pm 0.3) \times 10^{-9}$ m²/s; and ethane $D = (4 \pm 1) \times 10^{-10}$ m²/s.

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