

THE DC CONDUCTION MECHANISM OF ETHYLENE GLYCOL WATER SOLUTIONS

Dorota Nowak-Woźny — Tadeusz Mączka *

The effect of temperature and water content in ethylene glycol water solutions upon the electrical characteristics was studied in some details. The temperature was raised from room temperature up to about 380 K. It was suggested that the dependence between conductivity and temperature could be described by an exponential relation with the exponent depending on the potential barrier and temperature. It was supposed that the calculated potential barrier (0.2 eV to 1.1 eV) was associated with the motion of OH-groups.

Key words: DC conduction, potential barrier, charge transport, ethylene glycol

1 INTRODUCTION

Ethylene glycol and its water solutions are often used as a heat carrier in the industry. Ethylene glycol has good solubility in water. To obtain a solution with the best parameters, dielectric relaxation of ethylene glycol-water mixtures was performed over the whole concentration range at room temperature. The dielectric relaxation behaviour of mixtures of polar molecules under varying composition has attracted considerable interest because it helps formulating adequate models of liquid relaxation and also obtaining information about the relaxation process in the mixture [1–10]. These dielectric relaxation studies confirmed that the relaxation time of the system had a linear behaviour with the mole fraction of water in the mixture. It was also concluded that ethylene glycol (EG) molecules and surrounding water molecules formed EG-water clusters via hydrogen bonds and the nature of homogeneous and heterogeneous clusters changed with the water concentration in the binary mixture. Several attempts were made [11–21] to study the dielectric behaviour of various homologous series of alcohol mixtures with water and other polar solvents. At room temperature only one relaxation peak is observed for alcohol-water mixtures in the frequency range from 10 MHz to 20 GHz. The dielectric relaxation process of these systems is described by a sum of the Debye equation [22] and the Cole-Cole equation [23]. Though alcohol is the most popular material employed to study the hydrogen-bonding molecular liquids, it has been investigated systematically for only a few different alcohols. Experimental results of these binary systems are interpreted on the basis of a model of three kinds of cooperative domains coexisting in the mixtures.

The interest in the study of electric properties of glycol-water mixtures results from practical reasons. The

glycol-water mixture is a potential material for applications in electro heating devices. The goal of this work is to obtain glycol and water solutions with relatively low resistivity. Then dielectric parameters of different solutions of water and glycol were measured.

2 EXPERIMENTAL

The materials used for making the mixtures were pure ethylene glycol and deionized water.

All these experiments were carried out at temperatures in the range from 293 K to 373 K. The temperature was controlled thermostatically within ± 1 degree. Compositions and symbols for mixtures are presented in Table 1. Such mixtures allow a wide range of working temperatures as a technical point of view. The experiment was carried out in the unit presented in Fig. 1.

In the experiments, two rectangular plate electrodes were used. The rectangular plate electrodes were made of acid resistant steel. The electrodes are 0.105 m high and 0.062 m wide and are parallel to each other. The distance between the electrodes was kept by two Teflon plates. During the experiment the electrode surface temperature was measured in three points by thermocouple elements, type K, with a 0.25 mm diameter and LUTRON, type TM-906 A measurement equipment. The samples used in experiments were solutions of glycol (Table 1). The volume of each sample was 1 dm³. During the experiments, the air pressure was almost normal, the temperature was 295 K, and the relative humidity was about 45%. The resistance of electrodes and wires was more than 50 M Ω . The measurements of sample resistivity were carried out with a MOTTECH type MT 4080A RLC electronic meter. During the measurements the sample was under 1 V DC voltage and the resistance was measured after 60 s.

* Institute of Power Engineering and Fluid Mechanics, Wrocław University of Technology, 50-370 Wrocław, Wybrzeże Wyspiańskiego 27, Poland. E-mail: dorota.nowak-wozny@pwr.wroc.pl

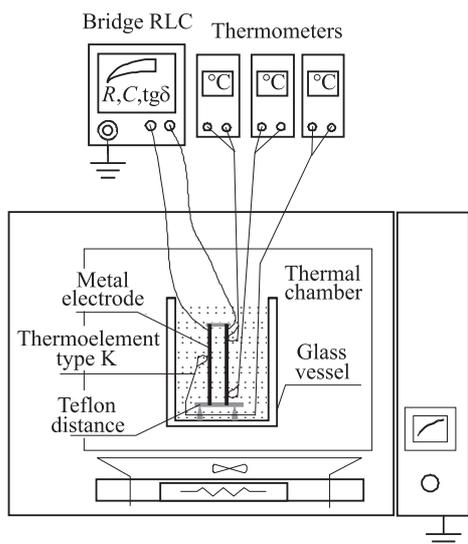


Fig. 1. The experimental unit.

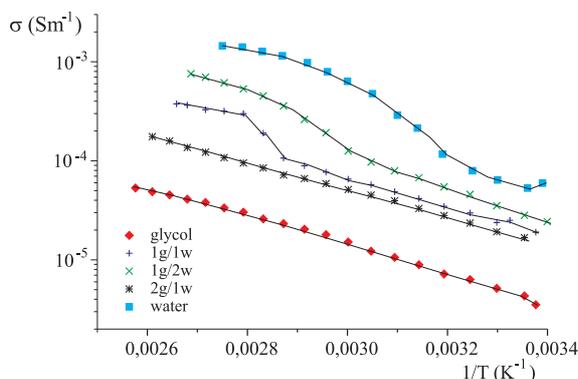


Fig. 2. Correlation between the conductivity and inverse of temperature for different mixtures.

Electrical measurements were made when the temperature in the thermal chamber was constant with accuracy ± 1 degree. The temperature was raised from room temperature up to the moment when intensive evaporation was observed.

3 RESULTS

Temperature variations of the conductivity of single samples (different ethylene glycol and water contents) were examined. The results are presented in Fig. 2. It is important that the conductivity was determined on the assumption that the measurement condition was always constant. For pure glycol the conductivity is smallest and for pure water it is largest in the whole temperature range. The results for $1_g/1_w$, $1_g/2_w$ and $2_g/1_w$ solutions are in the area between these two curves. The direction of resistivity variations with temperature differed quantitatively depending on the composition of the solutions. For pure water and $1_g/1_w$, $1_g/2_w$ solutions, three temperature ranges were observed in which the $\sigma(1/T)$ curves had different slopes. The shape of the resistivity

versus temperature curve depends on the water content. Interesting is the conduction mechanism. For pure glycol and $2_g/1_w$ solution the dependences between conductivity on a logarithmic scale and the inverse temperature are straight lines with nearly the same slopes (Fig. 2).

It is suggest that the dependence between the conductivity and temperature can be described by a relation like for liquid dielectrics [24–27]:

$$\sigma = \sigma_0 \exp(-E/kT), \quad (1)$$

where E is the potential barrier related to the conduction mechanism.

The values of activation energies were calculated and the results are shown in Table 2. For pure water three visible regions were observed but only for two of them the potential barrier was calculated. For the middle range the activation energy is equal to 0.6 eV and for the high temperature range it is 0.2 eV. For pure ethylene glycol and $2_g/1_w$ solution in the whole temperature range and for pure water and the other solutions in the lower and higher temperature ranges the activation energy is equal to 0.2 to 0.4 eV. For the transition region, observed only for pure water, $1_g/2_w$ and $1_g/1_w$, the potential barrier is 0.5 to 0.7 eV, 0.3 to 0.5 eV, and 1.0 to 1.2 eV, respectively.

The calculated potential barrier for pure water is by at least one order of magnitude lower than the dissociation energy.

It is difficult to explain why the potential barrier in the middle temperature range is higher than in the other regions. Probably there are two different conduction mechanisms. One mechanism is the motion of OH-groups and the other one is the motion of monomer units [27].

4 CONCLUSIONS

Measurements have been performed of the resistivity of different ethylene glycol water solutions. This work is a starting point for studying the conduction mechanism in ethylene glycol water solutions. These preliminary results obtained are promising and it seems that the direction of the research is right and proper.

REFERENCES

- [1] HOSAMANI, M. T.—FATTEPUR, R. H.—DESHPANDE, D. K.—MEHROTRA, S. C.: J. Chem. Soc. Faraday trans. **91** (1995), 623–626.
- [2] KUMBHARKHANE, A. C.—PURANIK, S. M.—MEHROTRA, S. C.: J. Chem. Soc. Faraday Trans. **87** (1991), 1569–1573.
- [3] SENGWA, R. J.—CHAUDHARY, R.: Polym. Int. **50** (2001), 433–441.
- [4] ROHMANN, M.—STOCKHAUSEN, M.—BUNSENGES, B.: Phys. Chem. **96** (1992), 677–681.
- [5] ROHMANN, M.—STOCKHAUSEN, M.: J. Chem. Soc. Faraday Trans. **88** (1992), 1549–1553.

Table 1. Solution compositions and their symbols.

Mixture content	Symbol
Pure ethylene glycol	EG
1 volume part of glycol and 1 volume part of water	$1_g/1_w$
1 vol. part of glycol and 2 vol. part of water	$1_g/2_w$
2 vol. part of glycol and 1 vol. part of water	$2_g/1_w$
Pure water	water

Table 2. The activation energy for different mixtures.

solution	Temperature range (K)	Activation energy (eV)
EG	296–393	0.3 ± 0.1
$2_g/1_w$	291–383	0.3 ± 0.1
$1_g/1_w$	296–348	0.3 ± 0.1
	348–358	1.1 ± 0.1
	358–376	0.2 ± 0.1
$1_g/2_w$	294–333	0.3 ± 0.1
	333–353	0.4 ± 0.1
	353–372	0.3 ± 0.1
water	296–340	0.6 ± 0.1
	340–370	0.2 ± 0.1

- [6] SHINYASHIKI, N.—SUDO, S.—ABE, W.—YAGIHARA, S.: *J. Chem. Phys.* **109** (1998), 9843–9847.
- [7] SHINYASHIKI, N.—YAGIHARA, S.—ARITA, I.—MASHIMO, S.: *J. Phys. Chem. B.* **102** (1998), 3249–3251.
- [8] SHINYASHIKI, N.—YAGIHARA, S.: *J. Phys. Chem. B.* **103** (1999), 4481–4484.
- [9] SENGWA, R. J.—KAUR, K.: *Polym. Int.* **49** (2000) pages 1314–1320).
- [10] SATO, T.—NIWA, H.—CHIBA, A.—NOZAKI, R.: *J. Chem. Phys.* **108** (1998), 4138–4147.
- [11] SENGWA, R. J.—CHAUDHARY, R.—MEHROTRA, S. C.: *Mol. Phys.* **109** (2001), 1805–1812.
- [12] BATEMAN, J. B.—GABRIEL, C.: *J. Chem. Soc. Faraday Trans.* **83** (1987), 355–369.
- [13] SATO, T.—CHIBA, A.—NOZAKI, R.: *J. Chem. Phys.* **110** (1999), 2508–2521.
- [14] SATO, T.—CHIBA, A.—NOZAKI, R.: *J. Chem. Phys.* **112** (2000), 2924–2932.
- [15] SATO, T.—CHIBA, A.—NOZAKI, R.: *J. Chem. Phys.* **113** (2000), 9748–9758.
- [16] SATO, T.—CHIBA, A.—NOZAKI, R.: *J. Mol. Liq.* **96-7** (2002), 327–339.

- [17] SATO, T.—CHIBA, A.—NOZAKI, R.: *J. Mol. Liq.* **101** (2002), 99–111.
- [18] SUDO, S.—SHINYASHIKI, N.—KITSUKI, Y.—YAGIHARA, S.: *J. Phys. Chem. A.* **106** (2002), 458–464.
- [19] CHAUDHAKI, A.—RAJU, G. S.—DAS, A.—CHAUDHAKI, H.—NARIN, N. K.—MEHROTRA, S. C.: *Ind. J. Pure Appl. Phys.* **39** (2001), 180–183.
- [20] PATIL, S. P.—CHAUDHAKI, A. S.—LOKHANDE, M. P.—SHANKARWAR, A. G.—HELAMBE, S. N.—ARBAD, B. R.—MEHROTRA, S. C.: *J. Chem. Engn.Data* **44** (1999), 875–878.
- [21] KHIRADE, P. W.—CHAUDHARI, A.—SHINDE, J. B.—HELAMBE, S. N.—MEHROTRA, S. C.: *J. Sol. Chem.* **28** (1999), 1031–1043.
- [22] DEBYE, P.: *Polar Molecules*, Chemical Catalog, New York, 1929.
- [23] COLE, K. S.—COLE, R. H.: *J. Chem. Phys.* **9** (1941), 341–345.
- [24] VOGEL, H.: *Phys. Z.* **22** (1921), 645.
- [25] TAMMANN, G.—HESSE, W.: *Z. Anorg. Allg. Chem.* **165** (1926).
- [26] FULCHER, G. S.: *J. Am. Soc.* **8** (1925), 339.
- [27] LEE, Y. C.—RATNER, M. A.—SHRIVER, D. F.: *Solid state Ionics* **138** (2001), 273–276.

Received 27 January 2006

Dorota Nowak-Woźny (Dr-Ing), was born in Wrocław, Poland. She has been studying solid state physics at the Wrocław University of Technology Faculty of Fundamental Problems of Technology; diploma thesis: the structure of thin Al films; PhD thesis: the precipitation hardening in doped NaCl crystals; 1983–1984: scientific worker at the Wrocław Oncology Centre; 1985–1998: research worker at The Low Temperature and Structure Research Institute Polish Academy of Sciences; 1998–2001: research worker at the Wrocław Department of Electrotechnical Institute; since 2001: research worker and lecturer at the Wrocław University of Technology, Faculty of Mechanical and Power Engineering, Institute of Heat Engineering and Fluid Mechanics; research activity: structure and microstructure defects and its influence on the electrical and mechanical properties.

Tadeusz Mączka was born in Bochnia, Poland, on February 28, 1972. He received the MSc degree in electrical engineering from the Wrocław University of Technology, Faculty of Electrical Engineering, Poland in 1997. Between 1996–2001 he worked as Laboratory Assistant, Engineer Technologist and head of Environmental Laboratory at the Electrotechnical Institute in Wrocław, Poland. At present he is assistant at the Wrocław University of Technology, Faculty of Mechanical and Power Engineering, Institute of Heat Engineering and Fluid Mechanics, Poland. His currently area of a scientific interest are: materials science and electrical technology, high voltage engineering and energy conversion - electrical process heating.