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The Rectifying Property of Polarized Barium Titanate

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Ternary System Ethyl Alcohol*n*-Heptane-Water at 30° C.

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OLUBILITY data for the ternary liquid system ethyl alcohol-*n*-heptane-water were obtained as part of a study of azeotropic distillation (4). Published data on this system (2, 3, 5) were incomplete, and the accuracy of these data was questioned. *n*-Heptane of high purity was not generally available when the earlier data were obtained.

MATERIALS

The ethyl alcohol used in this work was U.S.P., 200-proof (U. S. Industrial Chemicals, Inc.). The refractive index at

¹ Present address, C. F. Braun & Co., Alhambra, Calif.



Figure 1. Immiscibility Diagram at 30° C. of Ethyl Alcohol-n-Heptane-Water System



 $30\,^\circ$ C. was 1.3579. Pure grade *n*-heptane (Phillips Petroleum: Co.) was used. The minimum purity was 99 mole %. The refractive index at $30\,^\circ$ C. was 1.3835. Distilled water was used.



TABLE I. ETHYL ALCOHOL-*n*-HEPTANE-WATER SYSTEM

Fthyl	(*****	miseronity mile	4000 0.)	
Alcohol, Weight %	$n ext{-Heptane,} \\ ext{Weight} \\ \%$	Water, Weight $\%$	Density at 30° C.	Refractive Index at 30° C.
$\begin{array}{c} 0.00\\ 10.71\\ 28.31\\ 36.62\\ 45.65\\ 48.41\\ 59.90\\ 55.97\\ 64.95\\ 69.13\\ 72.92\\ 74.69\\ 69.13\\ 72.92\\ 74.69\\ 75.63\\ 68.62\\ 70.21\\ 75.63\\ 68.62\\ 70.21\\ 75.83\\ 49.88\\ 49.$	$\begin{array}{c} 100.00\\ 89.19\\ 78.86\\ 70.48\\ 61.90\\ 51.73\\ 48.91\\ 35.85\\ 40.46\\ 46.47\\ 29.44\\ 24.11\\ 17.85\\ 14.95\\ 10.10\\ 2.30\\ 2.26\\ 3.84\\ 6.69\\ 9.93\\ 0.21\\ 0.09\\ 0.90\\ 0.90\\ 0.16\\ 0.19\\ \end{array}$	$\begin{array}{c} \begin{array}{c} 0.10\\ 0.12\\ 1.21\\ 1.48\\ 2.62\\ 2.68\\ 4.25\\ 3.57\\ 3.21\\ 5.61\\ 6.76\\ 9.23\\ 10.36\\ 14.27\\ 29.08\\ 27.53\\ 20.79\\ 16.04\\ 13.69\\ 49.91\\ 74.89\\ 39.24\\ 99.84\\ 50.80\\ \end{array}$	$\begin{array}{c} 0.6750\\ 0.6843\\ 0.6931\\ \dots\\ \\ \end{array}$	$\begin{array}{c} 1.3835\\ 1.3807\\ 1.3779\\ \dots\\ 1.3779\\ \dots\\ 1.3669\\ \dots\\ 1.3655\\ 1.3642\\ 1.3629\\ 1.3620\\ 1.3621\\ 1.3620\\ 1.3622\\ 1.3622\\ 1.3612\\ 1.3612\\ 1.3622\\ 1.3622\\ 1.3582\\ 1.3582\\ 1.3582\\ \dots\\ 1.3620\\ \dots\\ 1.3322\\ \dots\\ \dots\end{array}$
		TIE-LINE D.	ATA	
	-	Ethyl Alcoho	1, %	
In heptane layer		In water layer		
	$2.9 \\ 3.2 \\ 5.0 \\ 5.0 \\ 5.5 \\ 14.5$		10.629.039.246.062.475.266.3	

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PROCEDURE AND RESULTS

Solutions of ethyl alcohol and *n*-heptane or water were prepared, and enough of the the third component was added to produce a cloud point. The solutions were made up by weight, using an analytical balance. The samples were held at $30.0 \pm 0.03 \text{ °C}$. in a constant-temperature bath, and were removed only for short periods to add the third component. Density and refractive index measurements were made on the samples at the cloud point.

The immiscibility curve for the ethyl alcohol-*n*-heptanewater system is shown in Figure 1. The data are given in Table I. The tie lines shown in this figure are for even percentages of water in the water-rich layer. The end compositions were taken from the correlation shown in Figure 3.

Densities were measured with a 5-ml. pycnometer, calibrated at 30° C. The wide and regular variation of density with composition makes it especially useful in the analysis of the ternary solutions (Figure 2). Density measurements were used for all analyses. The refractive index data were not used, but are included in Table I as an additional means of analysis. Tie-line data were determined by analyzing the two liquid phases of ternary mixtures. The two-phase mixtures were placed in a constant temperature bath at $30.0^{\circ} \pm 0.03^{\circ}$ C. and agitated periodically. The two phases were then separated and each was analyzed by utilizing the density-composition relation of Figure 2. The experimental results shown in Table I correlate by a method suggested by Bachman (1) (Figure 3).

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> The Rectifying Property of Polarized Barium Titanate

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When a high alternating voltage was applied across a wafer of polarized barium-titanate ceramic, a small direct voltage was observed across the ceramic. No such behavior was noted in wafers initially unpolarized. It is believed that the effect is caused by an asymmetrical charging action.

POLARIZED barium titanate ceramics appear to exhibit a rectifying action. While investigating the effect of high alternating voltages upon the amount of polarization present in the ceramics, it was found that a small direct voltage appeared to be present also. One circuit that was employed to observe this effect in more detail is shown in Fig. 1. The transformer was used to step up the alternating voltage to the value required and also to provide a return path for the direct current flowing in the galvanometer. The *RC* combination was, in effect, a low pass filter to attenuate the alternating voltage and to allow the direct current to pass.

The ceramic was in the form of a wafer with both flat surfaces silvered. It had been polarized by applying for one hour a direct voltage with the polarity shown in Fig. 1 and of approximately 20 kv per cm thickness of the wafer. For a resistance R of four megohms and with an rms alternating voltage of about 5.5 kv per centimeter across the wafer, the observed direct current indicated that the direct voltage across the wafer was about 0.6 volt per centimeter. The direction of the direct current flowing through the galvanometer is indicated by an arrow in Fig. 1 and indicates that an excess positive charge is present on the plate marked (-) of the ceramic wafer of Fig. 1. No direct voltage was observed when the polarized ceramic was replaced by an unpolarized piece.

At first it was thought that the alternating voltage was acting to reduce the remanent polarization of the ceramic and the stored energy of polarization was appearing as a charge on the plates of the wafer. This, however, would indicate that the direct voltage should have the opposite sign to that actually observed. Also the charge involved was of another order of magnitude greater than any of the measured values of the charges causing the remanent polarization. It is believed now that the polarization produces an asymmetrical charging effect in that more charge is stored when the current is flowing into the (-) terminal of the ceramic than when the current is flowing into the (+) terminal.



FIG. 1. Test circuit diagram for determining the rectifying properties of polarized barium titanate.

In a nonlinear capacitance such as barium titanate, the capacitance at any time may be defined as being proportional to the slope of the electric flux density D versus electric field intensity Ecurves which are also called hysteresis loops. It is well known that if the alternating voltage is high enough the capacitance of unpolarized barium titanate varies considerably over one alternating cycle but it has the same value for a positive voltage (when increasing, for example) as it does for a negative voltage (when decreasing). When polarized, however, this no longer appears to be true and the capacitance for a positive voltage may be quite different from the capacitance for the corresponding negative voltage. In polarized barium titanate the capacitance when current is flowing into the (+) terminal appears to be smaller than when the current is flowing into the (-) terminal. Thus a net positive charge appears on the (-) terminal. This seems apparent also from the asymmetrical hysteresis loops of polarized barium titanate since saturation appears on the loops when the voltage is positive on the (+) side of the wafer and thus lowers the capacitance in the (+) direction. Since the observed direct voltage is caused by this excess of charge, the magnitude of the voltage would probably depend upon, among other variables, the resistance of the discharge path and thus may be much higher if voltage measuring instruments of higher resistance were used. Possibly also voltage bucking or bridge circuits or better filters might enable better measurements to be made.

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