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Dielectric Properties of an Epoxy Resin

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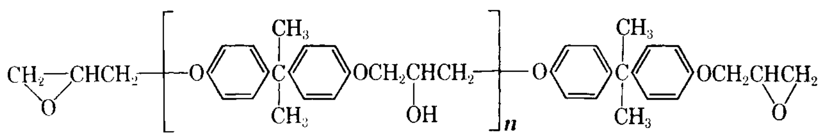
Synopsis

The dielectric properties of an unmodified epoxy resin with a wide range of hardener content were investigated over the frequency range 5 cycles/sec. to 50 Mcycles/sec and over the temperature range 12-95°C. In the frequency region below 1 Kcycle/sec., the loss is predominently high both in the pure resin and the pure hardener and, interestingly, is very much reduced when the two are mixed to obtain a plasticized product; the loss is at a minimum when the percentage of epoxy is about 10 and increases appreciably on both sides of the figure. At frequencies above 1 Kcycle/sec., a relaxation type loss is obtained in the hardener and the plasticized samples; pure resin again shows a predominently high loss, the maximum for it at room temperature being above 100 Mcycles/sec.

Introduction

The dielectric properties of an epoxy resin (I) (which is, according to the



manufacturers, a reaction product of bisphenol A and epichlorohydrin) with a wide range of hardener contents were investigated over the frequency range of 5 cycles/sec. to 50 M cycles/sec. and over the temperature range 12-95°C. The results obtained have already been reported briefly in a short communication[1] by the authors. In the present paper, these results are described in detail and an attempt is made to analyze them.

Experimental

The materials, Araldite B and Hardener 901, were obtained from CIBA of India Limited. The former contained some physical impurities which were removed by simple filteration process. Hardener 901 was used as received.

Disks of 2 in. diameter, 3-4 mm. thick, were prepared from plasticized resin containing 3.0, 5.0, 10.0, 20.0, 30.0, 40.0, and 50 wt.-% of the hardener. In each case, weighed amounts of resin and hardener were heated

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in a china dish and stirred well for a thorough mixing of the two. The molten mixture was then transferred to a mold designed in the laboratory, which was pretreated with silicone grease to facilitate the removal of the sample. The samples were cured by placing the mold in an oven adjusted to a temperature of about 125°C. for about 20 hr. After the mold had attained room temperature, the sample was removed, cleaned, and kept in desiccator overnight.

Samples of pure Araldite and Araldite with 3.0% hardener were not subjected to the curing treatment mentioned above. The hardener sample was made by pressing about 5 g. of material in an ordinary mold.

Dielectric loss measurements in the frequency range 5–160 Kcycles/sec. were made by the permittivity bridge method[2] and those in the range 200

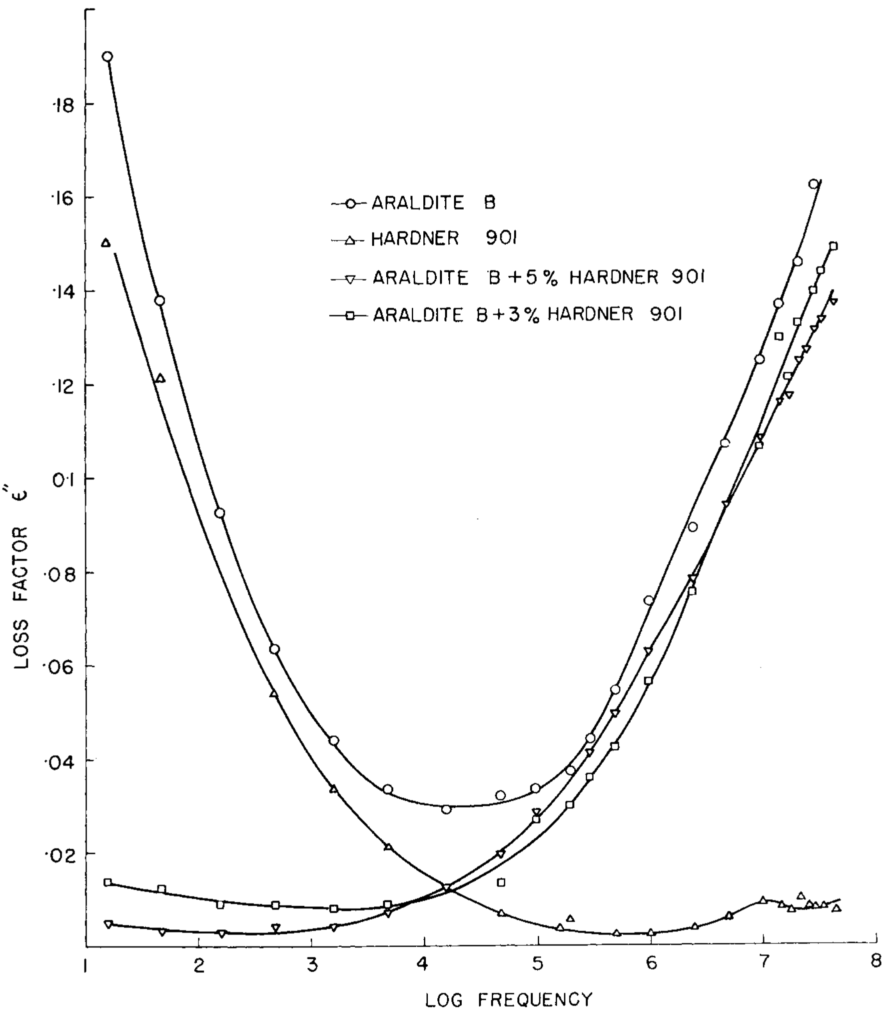

Figure 1.

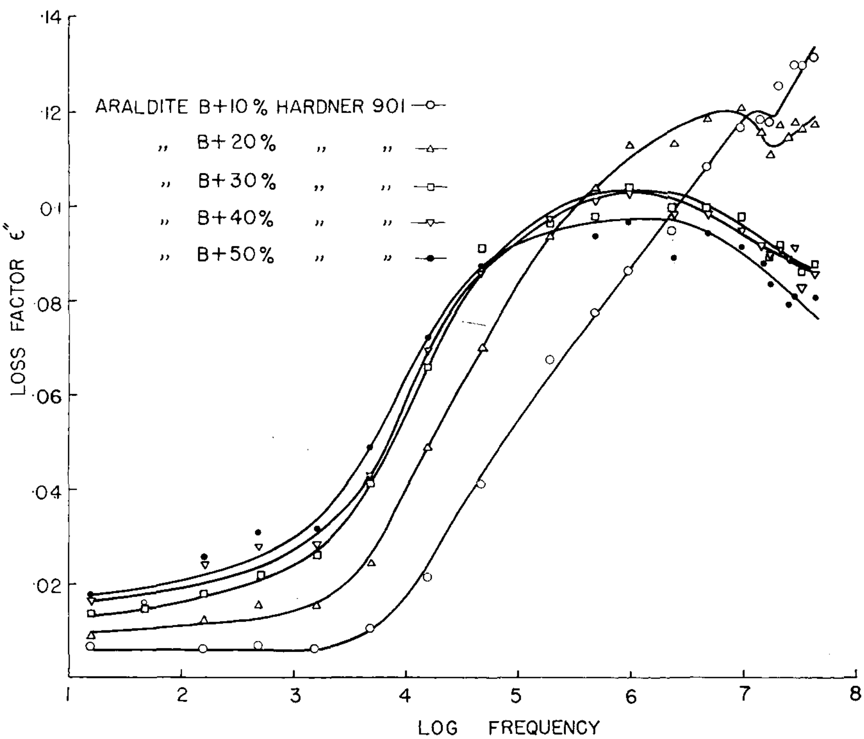

Figure 2.

Kcycles/sec. to 50 Mcycles/sec. were obtained by use of a Q-meter of the type 260-AP (Boonton Radio Corporation, USA) in conjunction with a dielectric sample holder, type 1690 (General Radio Co.).

Measurements of loss at different temperatures were made by use of a modified type 1690 sample holder designed and developed in the laboratory, in which the upper electrode formed a hollow cylinder and contained oil of high boiling point; the oil could be heated electrically and the temperature maintained constant to ±0.5°C.

Results and Discussion

In the frequency range below 1 cycle/sec., both pure Araldite and Hardener 901 showed high losses at room temperature (Fig. 1). The character of the curves indicates that the loss phenomenon is due to ionic conduction; this loss need not be the conventional Wagner-Maxwell type, because of the inhomogeneous nature of the ionic particles.[3]

In the plasticized products, the value of the dielectric loss in this frequency region is very much reduced. It was minimum in the product containing Araldite and hardner in the ratio of 10:1 (Figs. 1 and 2), and it increased appreciably on both sides of this ratio. Apparently, the explanation is that each component contained electrolytic impurities which re-

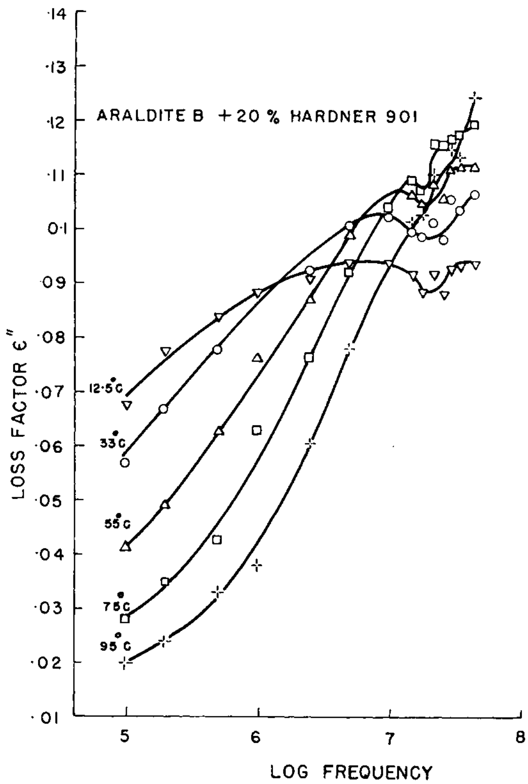

Figure 3.

acted on mixing to produce insoluble, nonconducting products, and the proportion of these in Araldite and hardener was of the order of 1:10.

Further, the epoxy resin (I) consists of complicated molecules and, possibly, the hydroxyl groups present in them provide the mechanism for the dielectric loss. But, such possibility is remote in Hardener 901, which consists of nothing but phthalic anhydride. Therefore, it is only the presence of ionic impurities to which the high loss effect is attributable.

In the high frequency region (1 Kcycle/sec. and above), pure Araldite shows high dielectric loss, the maximum value being beyond 100 Mcycle/sec.; the loss characteristics in this range are very much dependent upon a number of factors, such as the structure of the resin molecule, its chain length, and the nature of the dipole in the molecule.

In the plasticized product, a definite maximum of dielectric loss, representing dielectric absorption was obtained. As the percentage of hardener was decreased, the maximum tended to disappear, and with 5.0% or less of the hardener, the variation of dielectric loss with frequency was similar to that as in the case of pure Araldite. The increase in concentrations of hardener in the plasticized samples, on the other hand, only decreased the magnitude of the dielectric loss (Fig. 2).

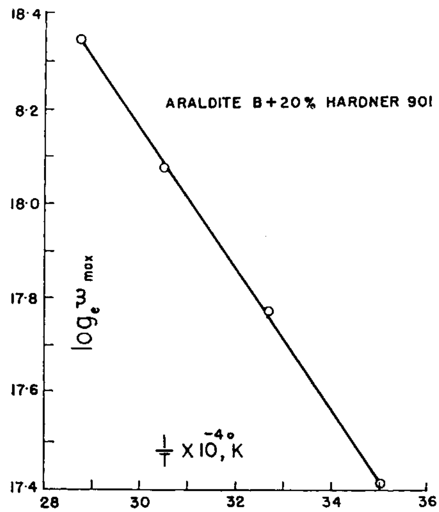

Figure 4.

The absorption characteristic which is indicated mildly in the loss curve for hardener (Fig. 1), was magnified considerably in the plasticized products. Fuoss[4] indicated that the presence of the plasticizer reduces the internal viscosity of the polymer–plasticizer system and that the response of the plasticizer dipoles in it appears independently. The present study provides another example of such a property.

The dielectric loss measurements for a particular plasticized sample containing 20.0% hardener were also recorded at different temperatures (Fig. 3). Assuming that the relation

$$\omega_{\text{max.}} \propto e^{-H/KT}$$

holds good in this case also, the value of H, the activation energy, was calculated (Fig. 4). This being approx. 3 kcal./mole, the mechanism of dielectric loss is clearly through the process of ion transfer.

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Résumé

Les propriétés diélectriques d'une résine époxy non modifiée contenant un pourcentage très variable de durcisseur ont été examinées dans un domaine de fréquence variant de 5 cycles/sec à 50 Mcycles/sec. et dans un domaine de température de 12 à 95°C. Dans la région de fréquence située en dessous de 1 Kcycle/sec; la perte des propriétés diélectriques dans la résine et dans le durcisseur est extrèmement importante et, ce qui est intéressant, est très reduite quand ils sont melangés pour obtenir un produit plastifié. La perte est minimum quand la pourcentage d'epoxy est environ égal à 10 et augmente appréciablement de part et d'autre de cette composition. Au delà de la région de 1 Kcycle/sec, une perte du type rélaxation est obtenue dans le durcisseur et dans les échantillons plastifiés; la résine pure montre de nouveau une perte extrêmement élevée, le maximum pour celui-ci à température ambiante se trouve au delà de 100 Mcycles/sec.

Zusammenfassung

Die dielektrischen Eigenschaften eines nichtmodifizierten Epoxyharzes mit einem in einem weiten Bereich liegenden Härtergehalt wurden im Frequenzbereich von 5 H_2/s bis 50 MH_2/s und im Temperaturbereich von 12 bis 95°C untersucht. Im Frequenzbereich underhalb 1 KHz/s ist der Verlust im Harz und im Härter vorwiegend hoch und wird interessanterweise bei Mischung beider zu einem weichgemachten Produkt sehr stark herabgesetzt; bei einem Prozentgehalt an Epoxy von etwa 10 erreicht der Verlust ein Minimum und steigt an beiden Seiten dieses Wertes stark an. Overhalb des 1 KHz/s-Bereiches wird im Härter und in den weichgemachten Proben ein Verlust vom Relaxationstyp erhalten; reines Harz besitzt wieder überwiegend hohen Verlust und das Maximum dafür liegt bei Raumtemperatur oberhalb 100 MHz/sek.

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