

FIG. 11 Variation of relaxation time filler concentration at 125 oC.

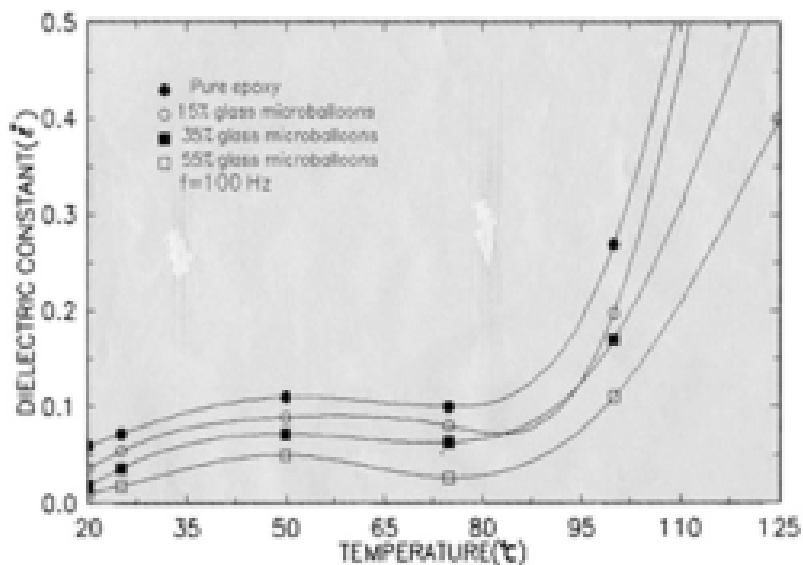


FIG. 9 Variation of dielectric loss with temperature at 10 kHz.

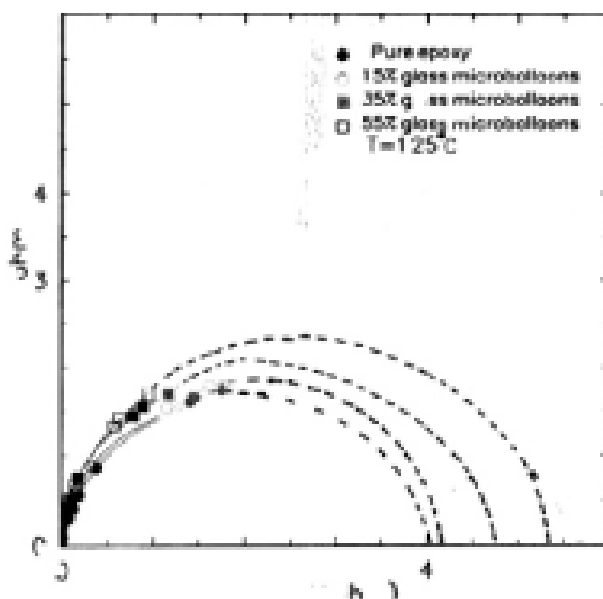


FIG. 10 Imaginary part of the impedance versus real part in the complex impedance plane at 125 oC.

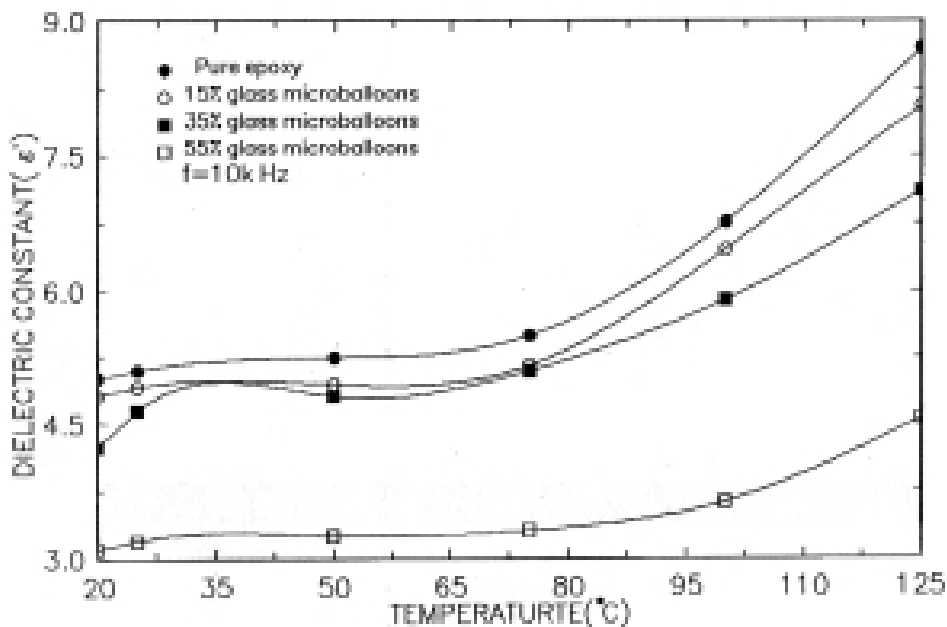


FIG. 7 Variation of dielectric constant with temperature at 10 kHz.

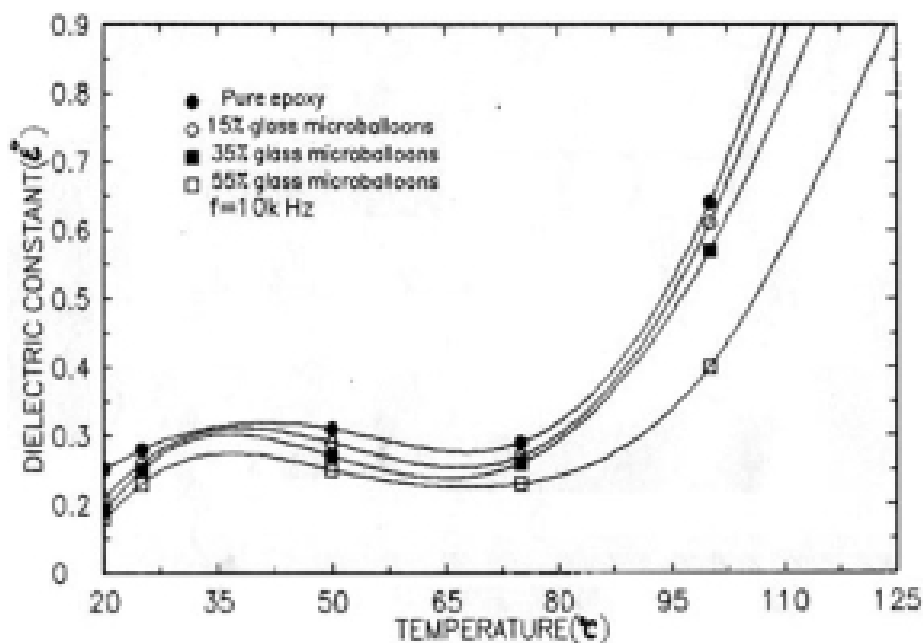


FIG. 8 Variation of dielectric loss with temperature at 100 Hz.

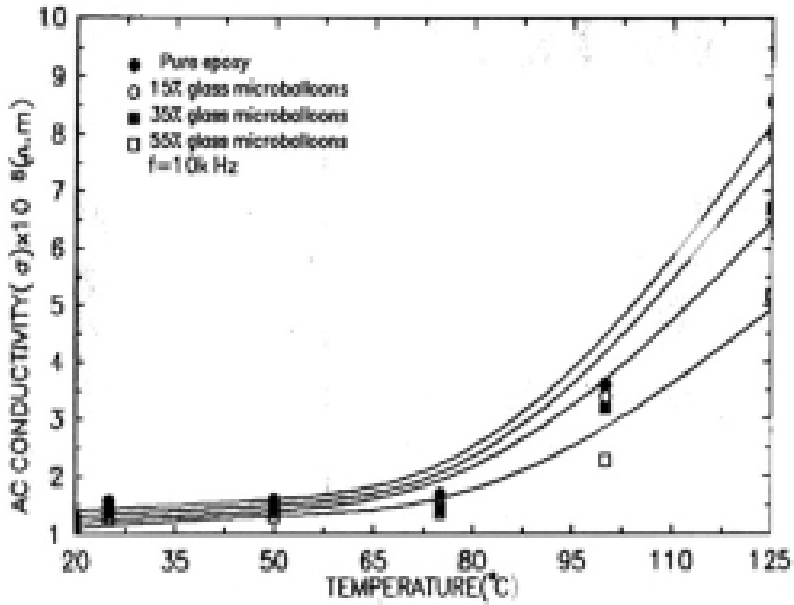


FIG. 5 Ac-conductivity versus temperature at 10 kHz.

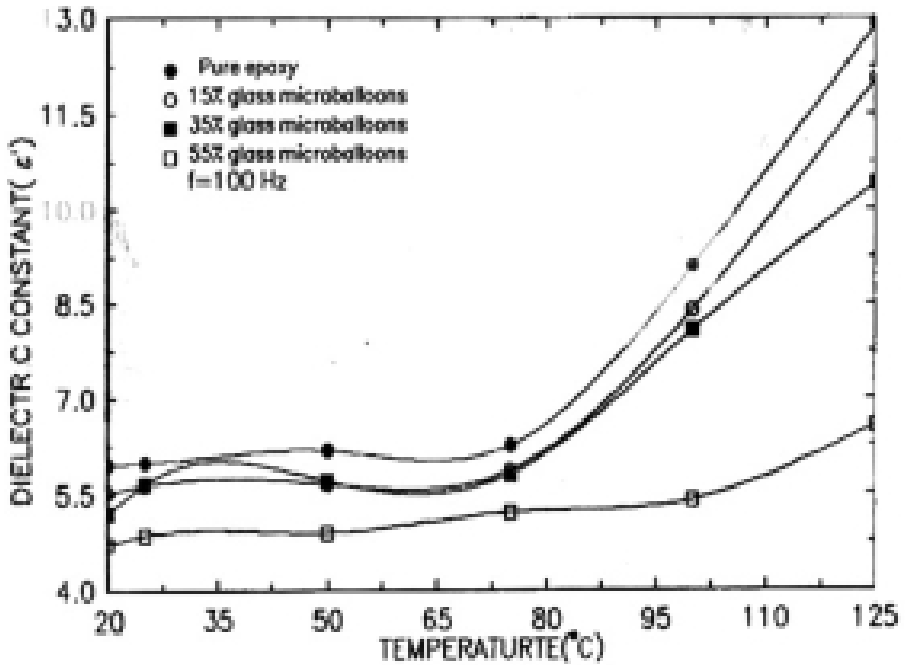


FIG. 6 Variation of dielectric constant with temperature at 100 Hz.

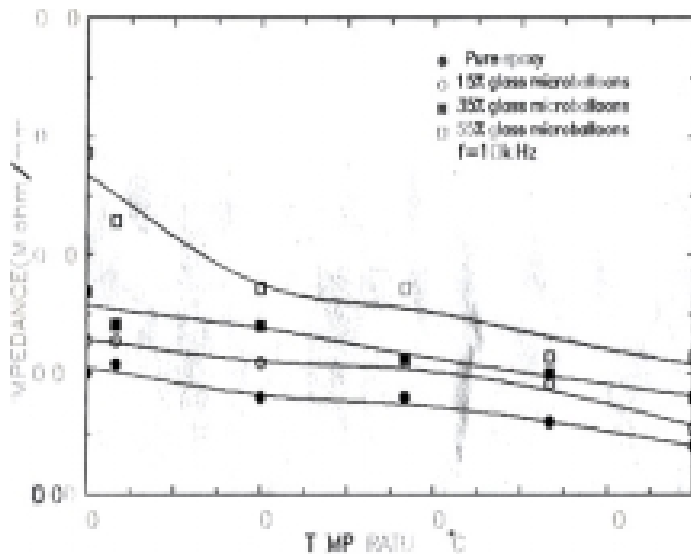


FIG. 3 Impedance versus temperature at 10 kHz.

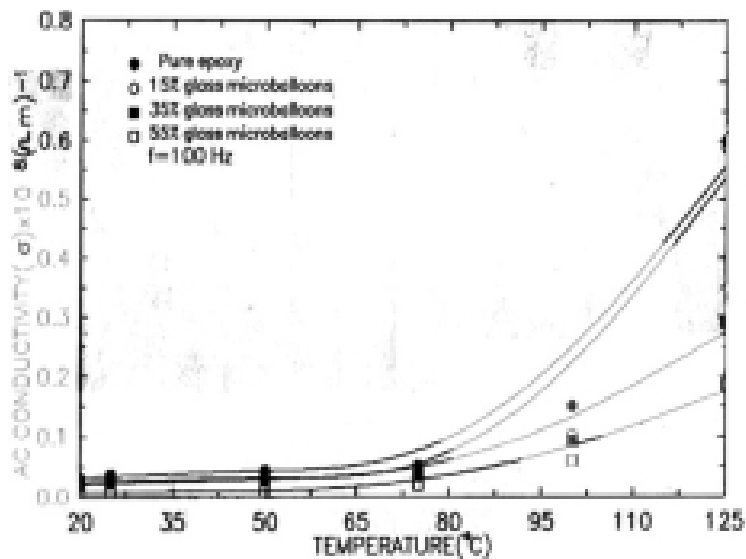
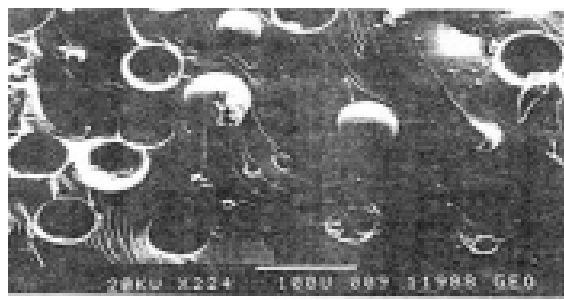
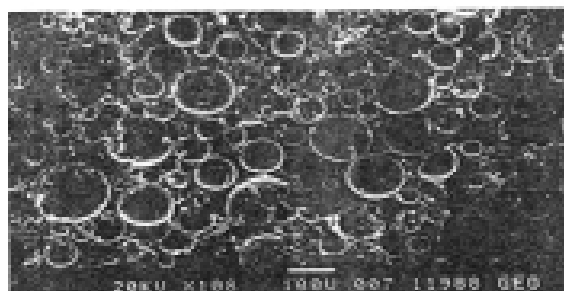


FIG. 4 AC-conductivity versus temperature at 100 Hz.



(a)



(b)

Fig. 1 SEM photographs for the epoxy composites  
 (a) 5 wt. % glass microballoons content.  
 (b) 10 wt. % glass microballoons content.

FIG. 1 SEM photographs for the epoxy composites.

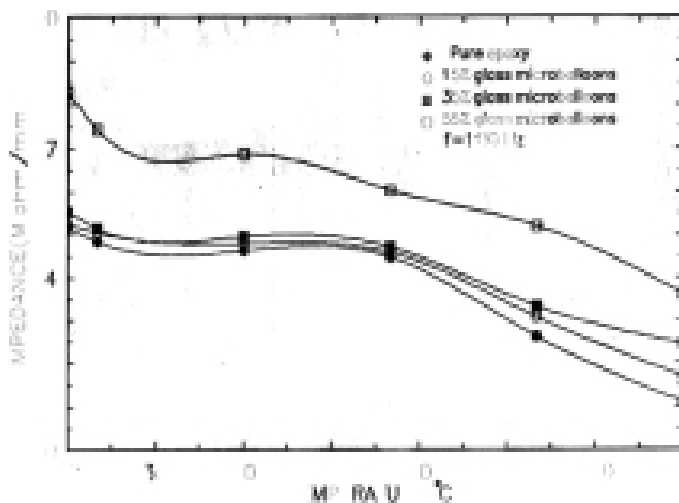


FIG. 2 Impedance versus temperature at 100 Hz.

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group- OH [8].

(Figure 10) shows plots for the imaginary component of the impedance  $Z_c$  against the real component  $Z_r$  at 125 oC. The plots yield distorted Cole- Cole circles, but it was not possible to construct such a pot at low temperatures, as  $\phi$  is close to  $-90^\circ$ , i.e.  $\cos(\phi) = 0$ . The possibility of constructing a Cole-Cole plot at 125oC ( $\phi$  at high temperature) indicates that the material is dominated by complex polarization mechanisms with different relaxation times. After extrapolating the lines to complete semi- circles, the relaxation time ( $\tau$ ) was determined from the plot of  $Z_c$  and  $Z_r$  using the relation ( $\tau = 1/\omega$ ), for different filler concentrations, where  $\omega$  is the angular frequency at maximum value of  $Z_c$  observed from figure 10.

The variation of the relaxation time ( $\tau$ ) with wt.% of filler content was shown in Figure 11. The relaxation time increases with increasing the filler content which indicates that the motion of the polymer chains is more restricted and hardened by the embedded filler glass spheres.

#### 4. Conclusion:

The dielectric results of the given epoxy-glass microballoons composite obtained between 20 and 125 oC reveal the following conclusions:

- 1- The impedance in general is decreases with increasing temperature above 80 oC due to thermal behaviour of the epoxy matrix near the glass transition temperature.
- 2- The AC-conductivity is enhanced at high temperature above 80 oC due to ionic and molecular mobility stimulated at high temperature ( $T > T_g$ ).
- 3- The dependence of dielectric constant and dielectric loss on temperature is attributed to ionic conductivity.
- 4- The complex impedance plane plots show spectra of relaxation times which imply that different conduction mechanisms are possible to take place at different temperatures.



the epoxy matrix. Other features of the composite morphology can also be drawn from the micrographs as the heterogeneous distribution due to mechanical mixing of glass microballoons with the epoxy resin. The thermal curing may displace the microballoons to stick to each other, especially at high filler concentration.

(Figure 2) shows the dependence of the AC impedance  $Z$  on temperature at frequency 100 Hz for different samples. A very weak dependence of  $Z$  was observed below 80 °C, while a strong one was observed above that temperature. Similar behaviour was noticed in (Figure 3) at frequency 10 kHz. This effect may be attributed to the thermal behaviour of the epoxy matrix above the glass transition temperature ( $T_g \approx 90$  °C) [7].

At low frequency  $T$  was more effective on  $Z$  than at high frequency, which may be due to a considerable increase in the ionic mobility, and structural defects of the composite are less pronounced.

The calculated AC conductivity ( $\sigma$ ) versus temperature is shown in (Figures 4 and 5) at 100 Hz and 10 kHz, respectively. The conductivity shows weak variation below 80 °C, while ( $\sigma$ ) was enhanced strongly above 80 °C which may be mainly due to the electron activation that increases rapidly with temperature, and due to ionic and molecular mobility stimulated at high temperature ( $T > T_g$ ) [2].

(Figure 6) shows the temperature dependence of the dielectric constant ( $\epsilon'$ ) at 100 Hz. The figure shows a weak growth below 80 °C, while the increase in ( $\epsilon'$ ) enhanced with temperature above 80 °C. This can be explained in terms of a large number of dipoles which are blocked at low temperature will be relaxed at high temperature. Similar behaviour was observed at frequency 10 kHz as shown in (Figure 7).

(Figures 8 and 9) show the plot of dielectric loss ( $\epsilon''$ ) versus temperature at frequencies 100 Hz and 10 kHz, respectively. The Figures indicate two peaks: one at 50 °C corresponds to ( $\epsilon''$ ) relaxation process and the other above 80 °C corresponds to ( $\epsilon''$ ) relaxation process [4] this peak was not formed completely since it goes to high temperatures above 125 °C. The ( $\epsilon''$ ) relaxation process associated with segmental motion or glass transition process occurs only close to  $T_g$ , and may be related to small chain movements. However, the ( $\epsilon''$ ) relaxation may be attributed to the relaxation of glyceryl unit-  $\text{O-CH}_2\text{-CH(OH)-CH}_2\text{-}$ , especially the local motion of the side groups in the resin hydroxyl

cooled in the oven to room temperature. Four disk-shaped specimens of 1.3, 1.1, 0.71 and 0.35 mm thickness and 20 mm diameter were machined from the composite sheets. The surface of test specimen was coated by thin gold layer by vacuum evaporation to serve as electrodes for electrical measurements using the ring method described elsewhere [10]. A suitable cell was designed to hold firmly the specimen under investigation.

## 2.2 Impedance Measurements:

The phase shift and the AC-impedance of the given composite were measured by a gain-phase meter. This instrument is capable of measuring directly the ration of the input to the output signal in dB and the phase angle ( $\theta$ ) in degrees as a function of frequency. The real component ( $Z_r$ ) and the imaginary component ( $Z_c$ ) of the complex dielectric constant ( $\epsilon^*$ ) are related to the impedance ( $Z$ ) and the phase angle ( $\theta$ ) as:

$$\begin{aligned} Z &= \sqrt{Z_r^2 + Z_c^2} \\ \theta &= \tan^{-1} \left( \frac{Z_c}{Z_r} \right) \end{aligned}$$

Where,  $f$  is the frequency,  $C_o$  ( $O/A/T$ ) is the capacitance of the electrodes,  $T$  is the specimen thickness,  $O$  is the permittivity of free space,  $A$  is the area of the disk.  $Z_c$  and  $Z_r$  are the imaginary and real components of the complex impedance ( $Z = Z_r - jZ_c$ ), respectively [6 ,7]. The AC electrical conductivity ( $\sigma$ ) was calculated from the relation:

$$\sigma = 2(f O \left( \dots \dots \right) \dots \dots (4)$$

## 3. Results and Discussion:

The SEM micrographs for the 5 and 55wt.% glass microballoons composite samples (Figure 1) show the textural and morphological evaluation of the composite as a function of filler content. It is clear that at low concentration, the microballoons are distributed in the matrix with no surface contacts between them, while at high concentration some contacts are existing between the adjacent glass microballoons. A small number of glass microballoons are pulled out from fractured surfaces which appears with dark spheres through

# ***TEMPERATURE DEPENDENCE OF ELECTRICAL PROPERTIES OF EPOXY- GLASS MICROBALLOONS COMPOSITE***

## **1. Introduction:**

For epoxy composites with tailored physical properties depending upon the application and the manufacturing process selected, a number of additives are often employed to provide specific procedures or end use properties. Modification of the electrical properties of epoxy composites has led to widespread applications, especially in the electronic industries and space instrumentation [6,9]. During the past few years, few publications have spread on the behavior of composite materials even that containing hollow microspheres [5,11,12]. As far as we know, no studies have been conducted to assist or characterize the dielectric behaviour of the given light, structure- composites. This paper is one of a series devoted to assess the mechanical and electrical behaviour of some advanced polymer composites with good performance [1,3].

The present paper essentially deals with the effect of temperature on the dielectric properties of epoxy composites with different filler concentrations at frequencies 100 Hz and 10 KHz.

## **2. Experimentation:**

### **2.1. Materials:**

The material used was epoxy- microballoons composite supplied by Shell company in U.S.A. The epoxy composite was prepared from the epoxy resin Epon 828 cured by V-40 and hollow glass microballoons spheres of size ranging from 10 to 180 (m and typical effective density 0.21 gcm<sup>-3</sup>. Sheets of different glass microballoons concentration (0,15,35, and 55 wt. %) were prepared by mixing the epoxy resin, curing agent and the microballoons. The mixture was stirred completely. The test samples were allowed to react at 25 oC for 24 hours and cured at 80 oC for 3 hours in an oven. The final step was to post cure the test specimens at 150 oC for two hours. Specimens were slowly

## ملخص

هذه الدراسة تتعلق بتأثير درجة الحرارة على بعض الخصائص الكهربائية للمتراكب ابو كسي \_ زجاج لعينات ذات تراكيز مختلفة من الحشوة التي هي عبارة عن كرات زجاجية صغيرة مملوءة بالهواء تتفاوت تراكيزها من صفر الى ٥٥٪ من الوزن، وذلك في مدى درجات الحرارة من ٢٠ م الى ١٢٥ م على الترددات، ١٠٠ هيرتز و ١٠ كيلوهيرتز. لقد وجد أن ثابت العزل الكهربائي يعتمد كثيراً على درجة الحرارة فوق ٨٠ م، كما اظهر الاعتماد على درجة الحرارة لمعامل الفقد عمليتي ارتخاء عند درجتي الحرارة ٥٠ م و ٨٠ م.

**Abstract:**

This study is concerned with the temperature dependence of some electrical properties of epoxy-glass microballoons composite containing 0, 15, 35, and 55% by weight glass microballoons. The temperature was varied from 20 oC to 125 oC at frequencies 100Hz and 10 KHz. It was found that the calculated dielectric constant showed a strong dependence on temperature above 80 oC . Two maxima occurred in the dielectric loss as a function of temperature plot. The low temperature peak around 50 oC was assigned to the (- relaxation process, and the high temperature peak began to appear above 80 oC to the (- relaxation process.

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PROPERTIES OF EPOXY- GLASS  
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