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POLYMER-CERAMIC COMPOSITES(U) UNIVERSITY COLL OF NORTH  
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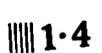
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Polymer - Ceramic Composites

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No. DAJ A45-86-C-0034

First and Second Interim Report

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Abstract

Composite films of PZT/PVDF have been prepared in which inclusions of PZT (ceramic) have been made in the matrix of PVDF (polymer). Preliminary observations show that a permittivity in excess of 100 may be obtained for such composites at low frequencies which is about an order of magnitude higher than that of the host polymer. The dielectric relaxation of this composite film conforms to that of the polymer (PVDF).

Introduction

Composite films containing ceramic (ferroelectric) inclusions in the matrix of polymer may possess high dielectric constants with good electrical breakdown strength and enhanced piezo- and pyroelectricity. Such composites are important in the application of thin film high energy density storage capacitor and piezo- and pyroelectric sensors. However, such materials are not yet commercially available and their electro-active properties are as yet not established.

It is our objective to prepare composite films with suitable ceramics (C) embedded in polymer (P) matrix to obtain as high as possible electro-active properties (ie high dielectric constant with good electrical breakdown strength and piezo- and pyroelectric coefficients. It is estimated that this work will require a time period of 24 months. The first 12 month period will cover studies of dielectric and electrical properties and the second twelve months will be occupied for the investigation of piezo- and pyroelectric properties of these composite films.



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## Experimental

PZT powder with an average grain size of 20 microns was obtained from Unilator Technical Ceramics and pellets of PVDF were provided by Solfay. These two materials were mixed in various concentrations using a rolling mill operated at 160°C. The composite hides were then pressed in a temperature controlled hydraulic press to thicknesses in the range 100 to 200 microns. The use of high quality chromium plates for pressing allowed the production of films with a good surface structure. Samples were thoroughly cleaned with ethyl alcohol in an ultrasonic bath to remove surface contaminants. They were then annealed in an oven for 72 hours at 80°C to condition them. Samples were then provided with vacuum evaporated aluminium electrodes of 0.1 micron thickness and 24mm in diameter. Dielectric measurements were performed with these samples in the frequency range of 10 Hz to 100 KHz using a General Radio Bridge (Model 1621).

## Results and Discussion

Figure 1 shows the experimental values of the relative permittivity of the composites as a function of the volume fraction of the composites. These data were obtained at a frequency of 1 KHz. The dielectric constant  $\epsilon$  of a two-phase system with spherical dispersions may be expressed thus[1]

$$\epsilon = \left[ \frac{2(1-X)\epsilon_1 + (1+2X)\epsilon_2}{(2+X)\epsilon_1 + (1-X)\epsilon_2} \right] \epsilon_1 \quad (1)$$

where X is the volume fraction of the inclusions and  $\epsilon_1$  and  $\epsilon_2$  are the dielectric constants of the polymer and the ceramics respectively for the two-phase system.

For the present system, the dielectric constant of the ceramic  $\epsilon_2 \gg \epsilon_1$  and the dielectric constant of the continuous polymer phase and the equation 1 may be simplified thus:

$$\epsilon = \left[ \frac{(1+2X)}{(1-X)} \right] \epsilon_1 \quad (2)$$

We refer to equation 1 as Model A and equation 2 as Model B. On figure 1 we have also plotted the calculated permittivity graphs using above equations. It may be observed that there is reasonable agreement with the experimental and the calculated values of  $\epsilon$  and that we need only consider the polymer when fabricating high permittivity composites.

Figure 2 shows the frequency dependence of the relative permittivities of PZT, PVDF and a composite of the two materials in the frequency range of 10Hz to 100KHz. In order to optimise the vertical scale we have multiplied the PVDF data by 10 and divided the PZT data by 10. It may be observed (figure 2) that there is little change in PZT permittivity over the four decades of frequency. On the other hand, the permittivity of PVDF increases by nearly 40% as the frequency is decreased from 100KHz to 10Hz. The dielectric behaviour of the composite material is similar to that of polymer alone and it may be observed that values of permittivity in excess of 100 may be obtained for the composite material at lower frequencies.

Figure 3 shows the frequency dependence of the dielectric losses in PZT and PVDF. The Alpha-C relaxation process in PVDF may be observed to peak at 80Hz, whilst a broad process is evident in PZT. Although the relative magnitudes of the loss processes are similar in these two materials, it should be pointed out that the loss process in polymer is more significant because it implies a loss tangent of 0.1 compared to 0.001 in PZT.

The frequency dependence of the loss processes in the composites is shown in figure 4. The behaviour is again similar to that of the polymer up to 50% PZT content. No peak may be observed in the 75% PZT case but this may be simply because of the increasing the ionic effect on the presence of an interfacial polarization. However, it may be significant that the actual level of dielectric loss increases as the volume fraction of PVDF decreases. This may be due to a decreased steric hindrance in the presence of a filler rather than some more specific interaction mechanism.

Present results indicate that the dielectric properties of PZT/PVDF composite films are governed by those of the PVDF polymer. Further work is in progress to investigate low frequency (1Hz - 10 Hz) dielectric behaviour of such composites.

#### References

1. W.R. Tinga, W.A.G. Voss and D.F. Blossey, J. Appl. Phys., 44, 3897 (1973).
2. K. Nakagawa and Y. Ishida, J. Polym. Sci; Polym. Phys. 53, 4328 (1982).

#### List of Diagrams

- Figure 1: Calculated and experimentally determined values of relative permittivity at 1KHz of PZT/PVDF composite for different volume fraction of PZT.
- Figure 2: Frequency dependence of relative permittivity of PZT, PVDF and PZT/PVDF composite.
- Figure 3: Frequency dependence of dielectric loss in PZT and PVDF.
- Figure 4: Frequency dependence of dielectric loss in PZT/PVDF composite for different volume fractions of PZT.

Figure 1: Calculated and experimentally determined values of relative permittivity of PZT/PVDF composite at 1kHz for different volume fraction of PZT.

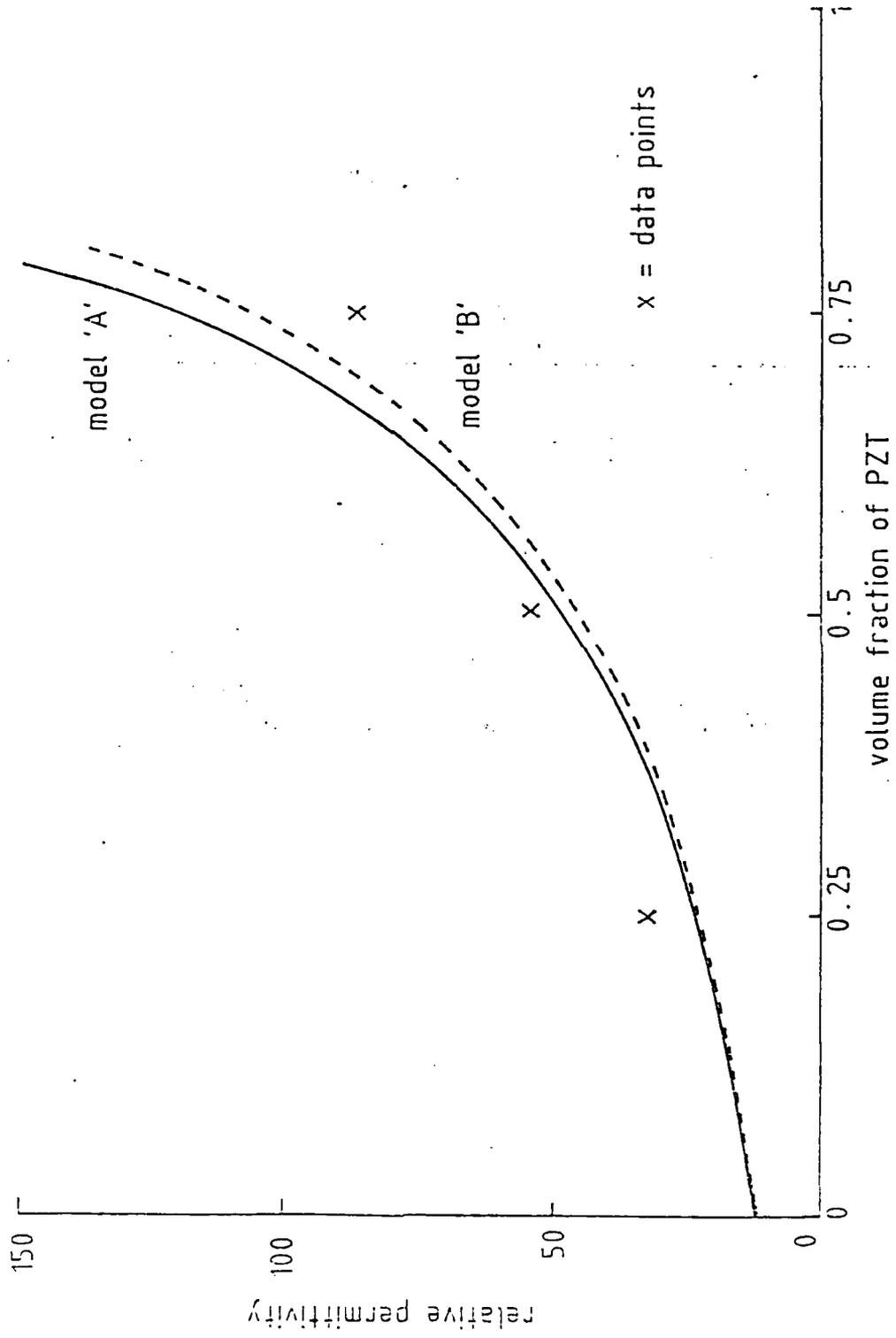




Figure 2: Frequency dependence of relative permittivity of PZT, PVDF and PZT / PVDF composite.

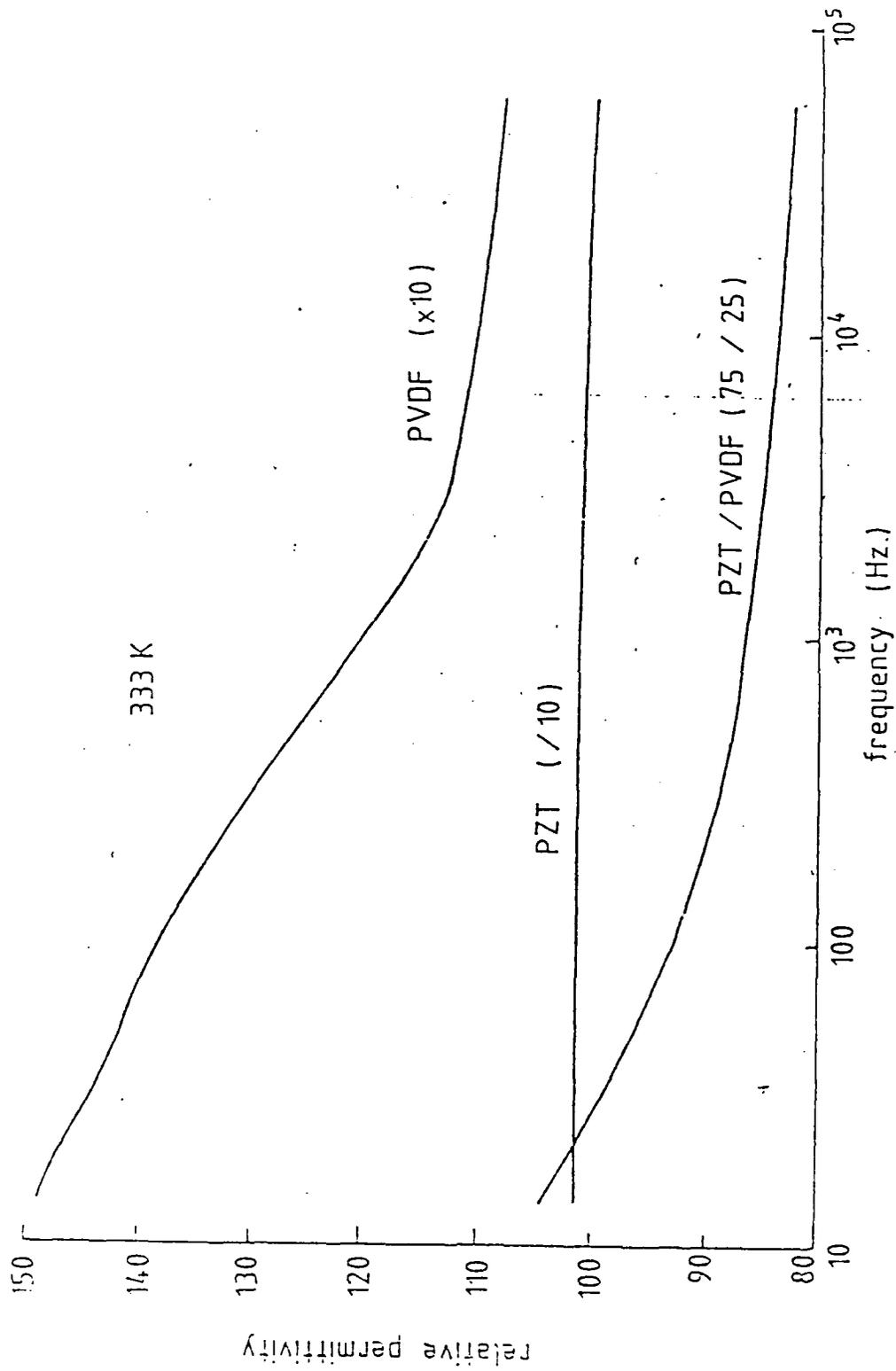


Figure 4 : Frequency dependence of the dielectric loss in PZT/PVDF composite for different volume fractions of PZT.

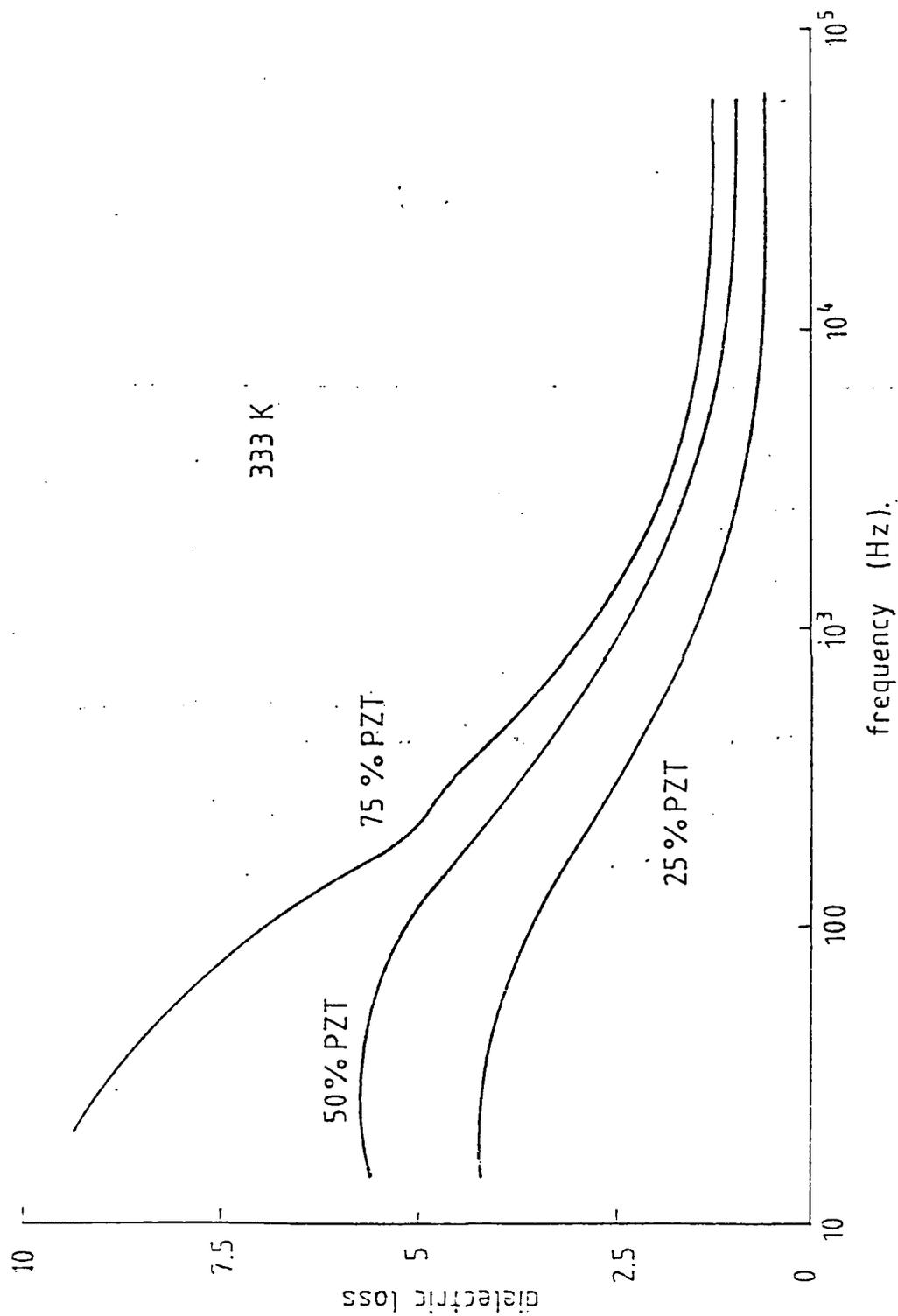
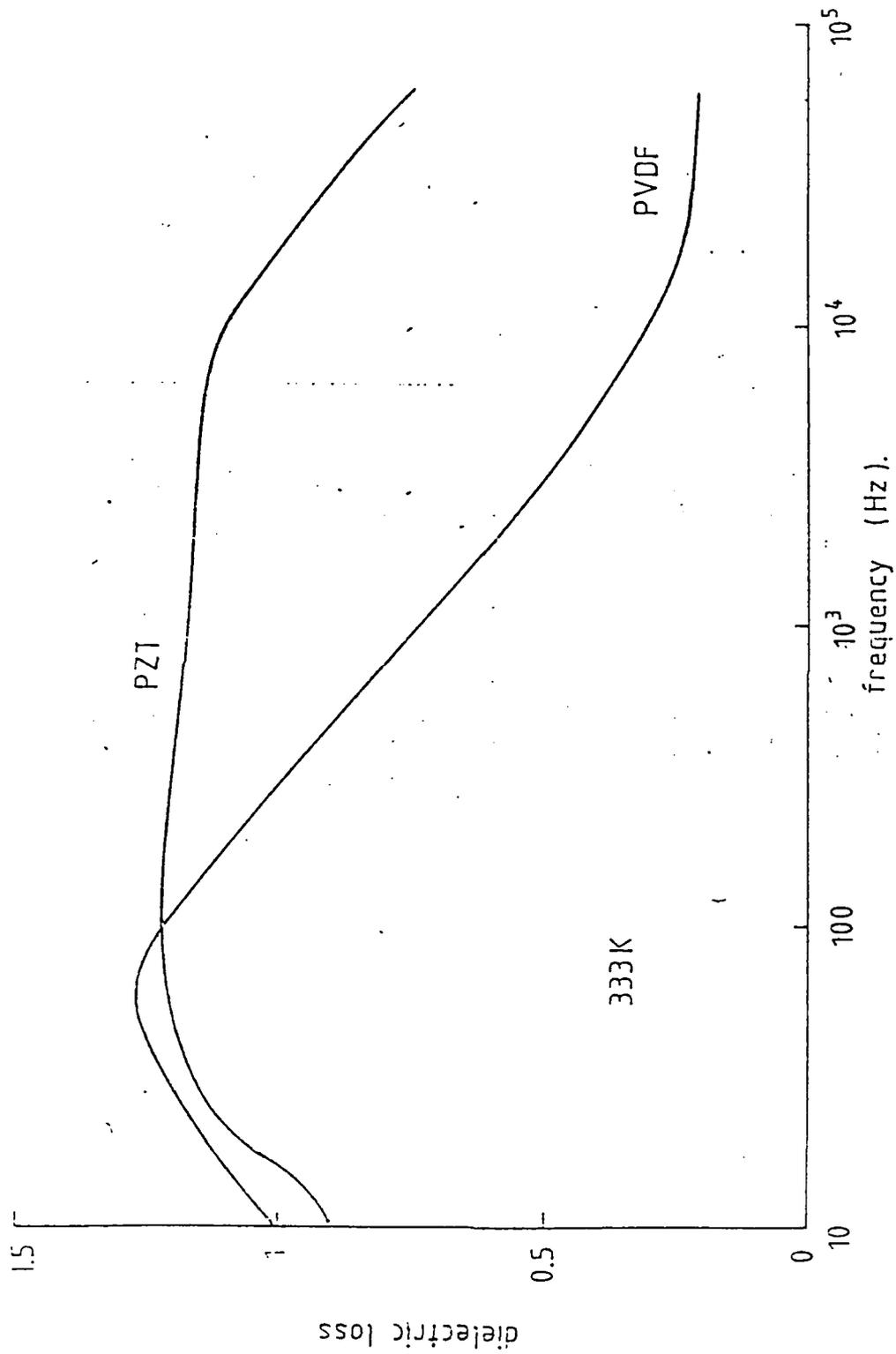


Figure 3 : Frequency dependence of dielectric loss in PZT and PVDF.



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