

## DIELECTRIC PROPERTIES OF ISATOMALONONITRILE

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The paper reports dielectric measurements carried out for isatomalononitrile at different temperatures (346 to 383 K) and various frequencies (50 to  $5 \times 10^5$  Hz). Kramers-Kronig relations are used to ascertain the magnitude of the dc conductivity and to enhance the reliability of dielectric measurements. The analysis of the results shows that there exist certain very slow dielectric phenomena in isatomalononitrile. The frequency dependent conductivity of isatomalononitrile increases approximately linear with frequency. The frequency exponent has the value of 0.5, independent of temperature.

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The dielectric properties of solids can be described by their complex capacitance defined by the expression

$$C^*(\omega) = C' - iC'' = C(\omega) - i \frac{G(\omega)}{\omega}, \quad (1)$$

where  $C'$  and  $C''$  are the real and imaginary components of the complex capacitance;  $C(\omega)$  is the ordinary capacitance,  $G(\omega)$  is the effective alternating-current conductance and  $\omega$  is the angular frequency.

The complex dielectric permittivity  $\epsilon^*(\omega)$  is related to  $C^*(\omega)$  by a geometrical factor and the corresponding expression for the complex dielectric susceptibility  $\chi^*$  may be obtained from  $\epsilon^*(\omega)$  by subtracting the dc contribution  $\sigma_0/\omega$  and the "high frequency" permittivity  $\epsilon_\infty$  [1]

$$\chi^*(\omega) = \chi'(\omega) - i\chi''(\omega) = \epsilon'(\omega) - \epsilon_\infty - i \left[ \epsilon''(\omega) + \frac{\sigma_0}{\omega} \right], \quad (2)$$

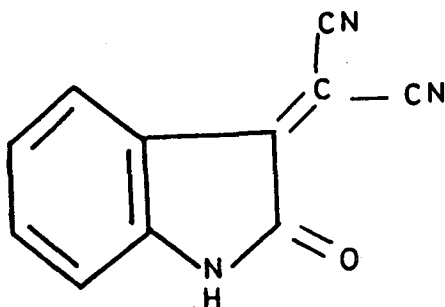
where the real and imaginary components of  $\chi^*(\omega)$  and  $\epsilon^*(\omega)$  are Kramers-Kronig (KK) compatible.

In the case of "universal" form of frequency dependence of both  $\chi'(\omega)$  and  $\chi''(\omega)$  on  $\omega^{n-1}$ , where the value of  $n$  being in the range of (0,1), their ratio is independent of frequency [1]

$$\frac{\chi''(\omega)}{\chi'(\omega)} = \cot \left( \frac{n\pi}{2} \right). \quad (3)$$

In view of the promising importance of isatin derivatives and their related compounds in biological systems [2-4] and in continuation to our previous work [5-6] and due to the scarcity of literature on the dielectric properties for this compound, in the present paper we report the dielectric properties of isatomalononitrile as a function of both temperature and frequency.

Isatomalononitrile was prepared by adding an ethanolic solution of malononitrile, to a solution of isatin in absolute ethanol (1 mol). The reaction mixture was heated under reflux for 1h. The solution was then reduced to a small volume by evaporation and left to cool whereby isatomalononitrile separated and precipitated. The compound was filtered off, washed with ethanol, dried and preserved in a desiccator over  $P_4O_{10}$ .



Isatomalononitrile or 3,3-dicyano methylene isatin.

The powder was compressed in a die under a pressure of  $9.8 \times 10^8$  Pa in the form of pellets of 10 mm in diameter and about 1-4 mm thickness. Silver paste was painted on the major faces of each pellet as electrodes. The dielectric measurements were done as a function of temperature in the frequency range 50 to  $5 \times 10^5$  Hz using impedance meter BM 507. Measurements were made in a cell specially designed to minimize the effect of stray capacitance. The temperature was measured by means of a copper constantan thermocouple mounted in the close proximity to the specimen of interest.

Figure 1 shows the results for isatomalononitrile pellets plotted as the real and imaginary components  $C'$  and  $G/\omega$  of the complex capacitance as a function of frequency and temperature. The real part ( $C' - C_\infty$ ) is also shown in Fig. 1, and was obtained by subtracting a suitable value of  $C_\infty(T)$  for each temperature to obtain KK compatible behaviour with the imaginary part. The KK compatible  $C'$  curve, for each temperature, is a straight line with a distance between ( $C' - C_\infty$ ) and  $C''$  curves given by  $\log \cot(n\pi/2) = 0$ , characterised by the value of  $n \approx 0.5$ . The exponent  $n$  remains constant, within the experimental error with increasing temperature. Below  $10^3$  Hz the slope of the  $G/\omega$  curve is found to be about -1, which gives the value of  $n$  close to zero and  $C' - C_\infty$  and  $C''$  curves are not KK compatible. This may be due to the dc phenomena.

At the lowest temperatures shown in Fig. 1, 346 K, a marked dispersion of loss and a corresponding slight dispersion of  $C'(\omega)$  at lower frequency becomes evident and this increases steadily with rising temperature. It may be assumed

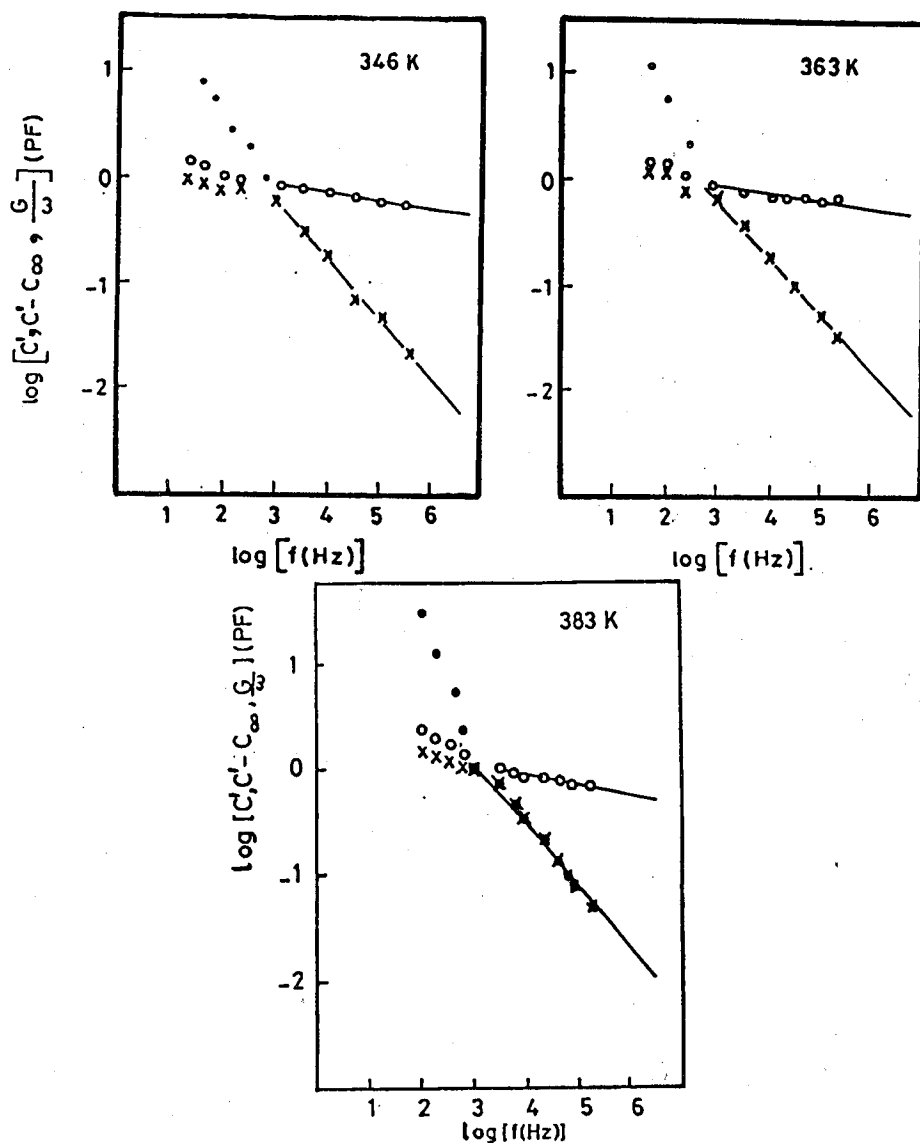


Fig. 1. Dielectric response  $C'(\omega)$  (o),  $G/\omega$  (●) and  $C''(\omega) - C_\infty$  (x) of an isatomalononitrile pellet taken over a range of temperatures rising from 346 to 383 K.

temperature. Below  $10^3$  Hz the slope of the  $G/\omega$  curve is found to be about  $-1$ , which gives the value of  $n$  close to zero and  $C' - C_\infty$  and  $C''$  curves are not KK compatible. This may be due to the dc phenomena.

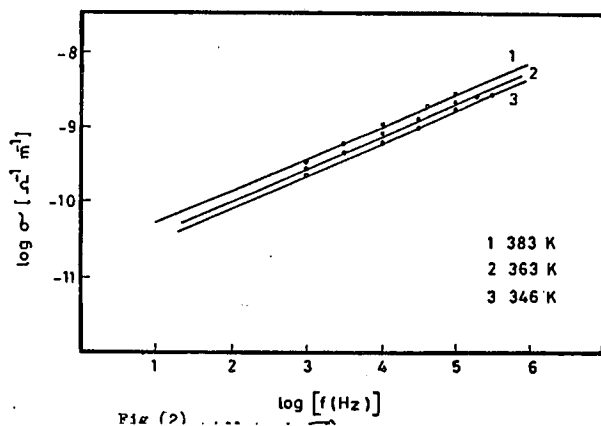


Fig. 2. The frequency dependence of the conductivity of an isatomalononitrile pellet for a range of temperatures.

rise to the low-frequency dispersion [7].

The frequency dependence of the conductivity of isatomalononitrile pellets in the high frequency range is shown in Fig. 2 for a range of temperatures. The low temperature data show a relatively "conventional" power law relation ( $\sigma \propto \omega^n$ ) with  $n \approx 0.5$ .

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