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Dielectric Studies of Some Indian Granite Samples

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Abstract:Granite samples were collected from various places of South India. Chemical analysis of these granite samples were investigated using X-ray fluorescence spectroscopy. The major contents observed are SiO_2, Al_2O_3 and Fe_2O_3. Larger grain size is observed in FESEM micrographs. This indicates longer time grown granites. Measurement of dielectric constant (ε) and loss (tanδ) have been carriedout on granite samples as a function of frequency from 1Hz to 5MHz and temperature from300K to 725K. An effective and alternate approach is adopted for investigating dielectric relaxationbehaviour viz. electric modulus spectroscopy. This includes fitting of dielectric response to Bergmann frequency domain function. The average activation energiesevaluated from these fitsare E_1=0.34eV and E_2=0.51eV. The results are discussed.

Keywords:dielectricconstant; dielectric studies;granites; electric modulus.

I. Introduction

In recent years, geological materials have been the subject of frequency dependent dielectric measurements over wide frequency ranges due to their significant applications in the Earth and Planetary Sciences and Technology developments[1-6]. The dielectric properties of rocks are primarily a function of mineralogy, frequency,water saturation, porosity,rock texture,and component geometry and electrochemical interactions. Dielectric dispersion studies in low frequency region help to understand the behaviour of induced polarisation in the materials[7],while high frequency studies are useful in planning ground penetrating radar surveys[8-10], in microwave remote sensing of the earth's geology of these materials and calibration of time domain reflectometrymeasurements[11-14].

Granite is a plutonic igneous rock because it is formed due to solidification of magma at greater depths. Earlier studies confirmed that rocks and minerals show the dielectric dispersion in low frequency region [15-17]. Few attempts were made to determine the dielectric behaviour of granite samples in low frequency region andalso at microwave frequencies[18-19]. Sengwa etal [2] studied the dielectric dispersion of Indian granites available in various locations of India in the frequency range 0.10-100 kHz and also at microwave frequency i.e 10.1 GHz at room temperature and they revealed that all dry granite samples showed dielectric dispersion in the low frequency range. Mostafa et all [3] studied the temperature dependence of electrical resistivity of granite samples and also revealed the behaviour of electrical resistivity of granite samples collected from Egypt have two conduction regions and their electrical resistivity exhibit a rapid decrease with the increase of temperature.However no systematic dielectric measurements were available for both as a function of frequency and temperature. Therefore an attempt has been made to report a systematic dielectric behaviour by choosing three popular variety granite samples available in different areas of Andhra Pradesh and Karnataka states through impedance measurements.

II. Experimental Details

The granite samples were collected from Karimnagar and Ongle districts of Andhra Pradesh state and Hasan district of Karnataka from South India. The samples were powdered to a required size and chemically analysed the composition using Philips PW2440 microprocessor controlled sequential X-ray fluorescence (XRF) spectrometer. The micrographs of the granite samples were taken using field emission scanning electron microscope (FESEM, CARL ZEISS ULTRA 55).The dielectric measurements were carriedout in the frequency range,1Hz to 5 MHz and temperature range,300K to 725K using a NOVOTEL ALPHA–A High frequency Analyser.Thin sections of granite samples (coated with silver paint) of thickness~1.6mm and area 180mm^2 were used for this purpose.

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III. Results and Discussion

A. X-ray fluorescence study

The composition of the samples obtained from XRF study has been tabulated (Table-1). The major contents observed in thesamplesare SiO_2, Al_2O_3 and Fe_2O_3.

TABLE I
Chemical Analysis (%) of the Samples using XRF

Sample	Place	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	Sum
Hasan Green (a)	Hasan Karnataka	49.05	10.44	21.91	0.15	3.38	7.23	2.82	1.35	1.87	0.32	98.52
Black Pearl (b)	Ongole Andhra Pradesh	55.28	15.75	11.91	0.1	0.88	4.04	2.68	6.46	1	0.68	98.78
Cat's Eye (c)	Karimnagar Andhra Pradesh	67.26	13.41	5.91	0.05	0.97	1.94	3.37	4.7	0.48	0.22	98.31

B. Microstructure:

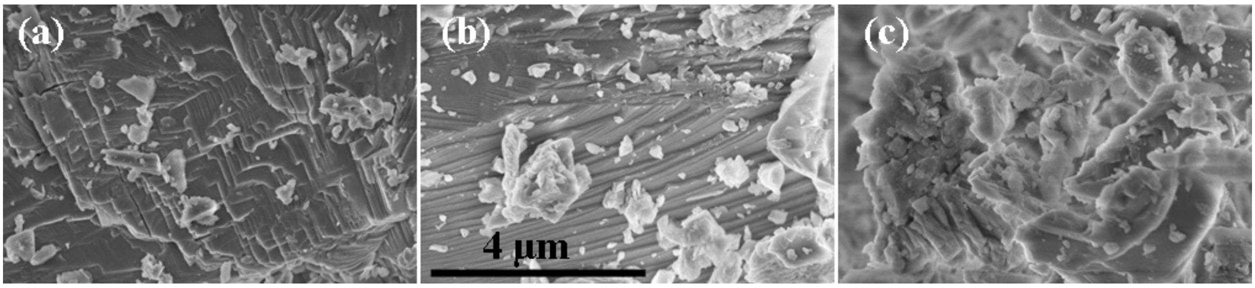

Fig.1 FESEM micrographs of the granite samples (a),(b) and(c).

Fig. 1 shows FESEM micrographs of the samples. Larger grain size is observed in the FESEM micrographs. This indicates longer time grown grain structure in the granite samplesand the continuity indicates more crystalline phase.

C. Dielectric constant (ε^1, loss($\tan\delta$) andε^{11})

Dielectric studies viz.ε^1,ε^{11} (real and imaginary parts of dielectric constant respectively)and $\tan\delta$as a function of frequency at different temperatures are presented for three samples (a),(b) and (c) in figures 2 to 4. The figures are shown in two different temperature regions separately viz. low temperature region (region I) and high temperature region (region II).

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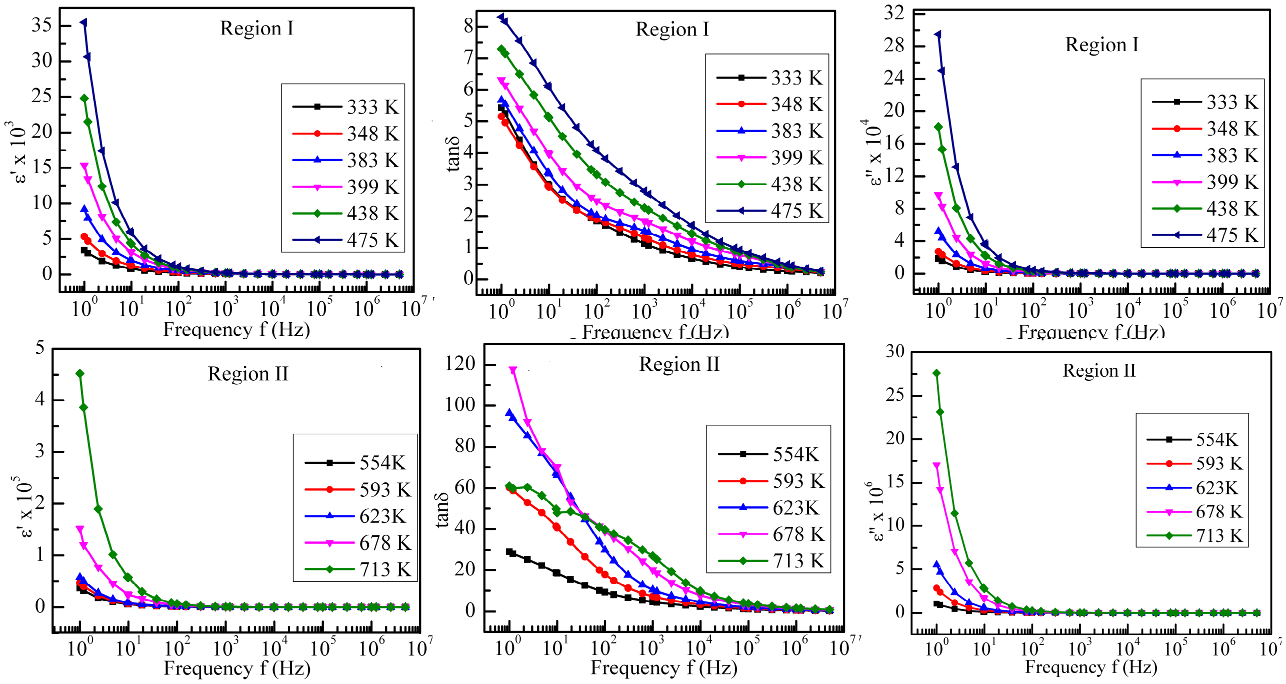

Fig. 2.ε^1, tanδ and ε^{11}as a function of frequency of sample (a) for two temperature regions

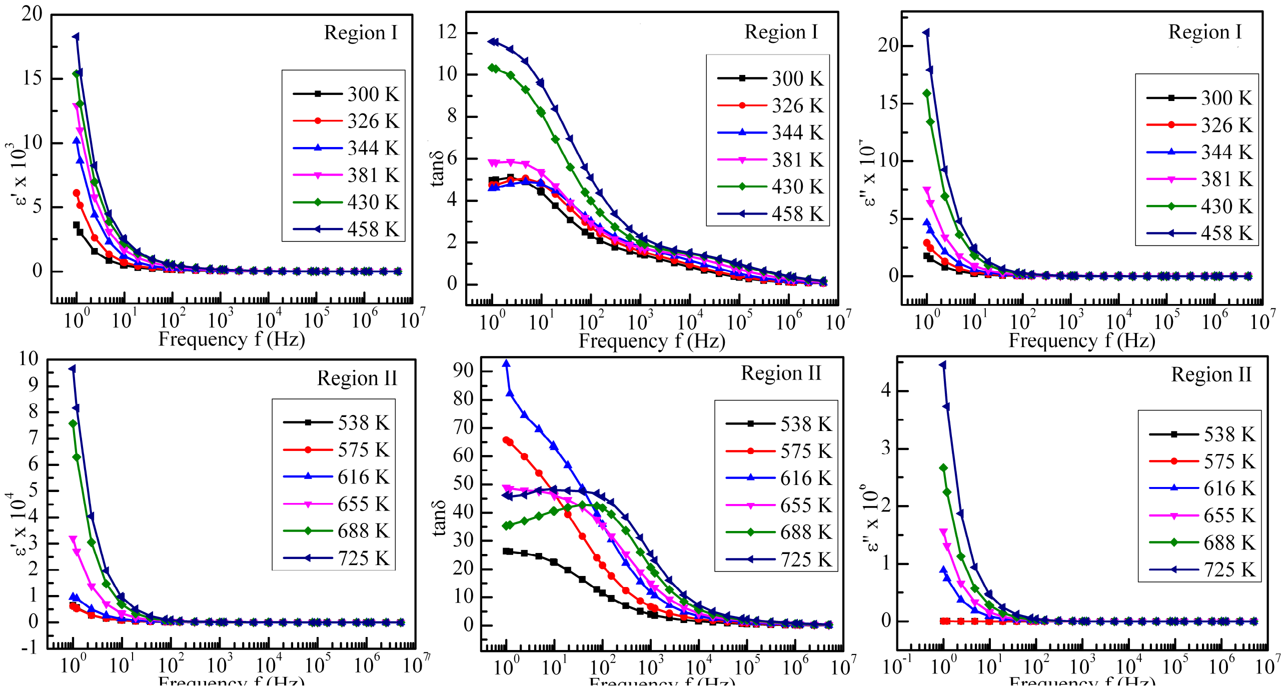

Fig. 3.ε^1, tanδ and ε^{11} as a function of frequency of sample (b) fortwo temperature regions.

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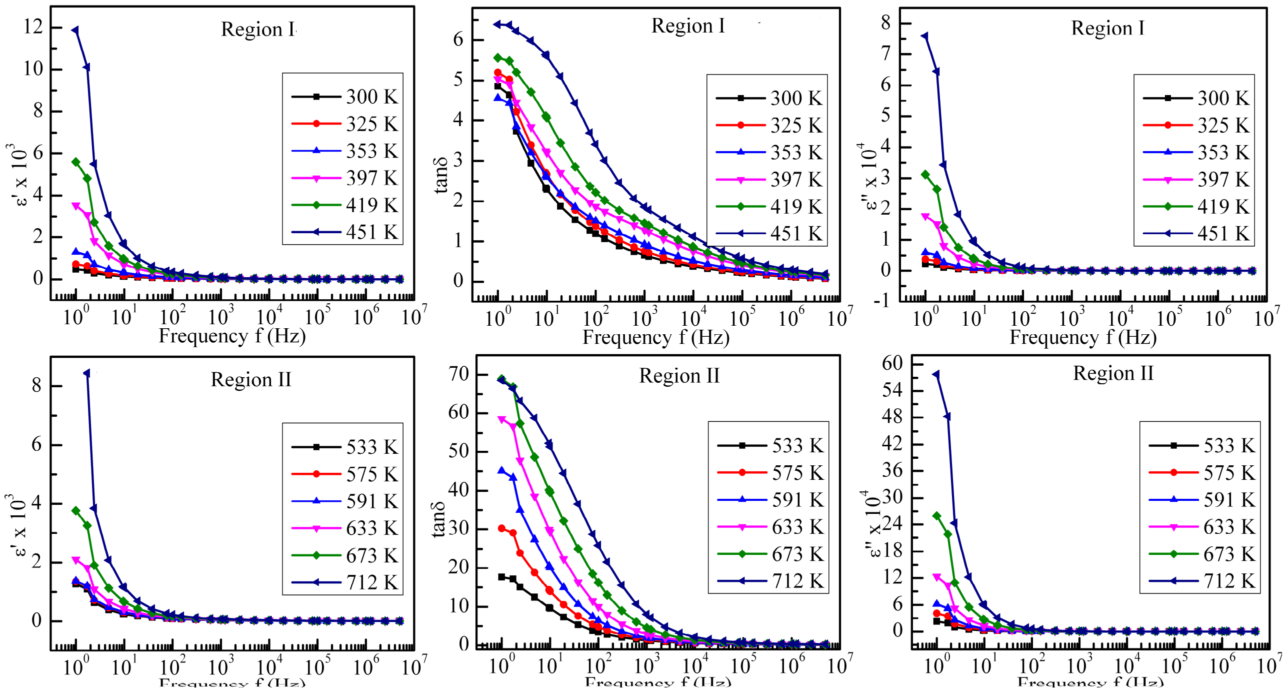

Fig. 4. ε^1, tanδ and ε^{11} as a function of frequency of sample (c) for two temperature regions

It can be seen from figures 2 to 4 that for given granite sample ε^1 is high at lower frequencies and decreases with increasing frequency. At higher frequencies ε^1 is found to be both frequency and temperatureindependent, while at low frequencies below 1kHzthe variation is temperature dependent. Similar variation is found incase of tanδ and ε^{11}also but in different order of magnitudes. The high value of ε^1 at low frequencies is believed to be due to space charge polarisation which arises due to grain boundaries and grain imperfections [2]. The same would be negligible at high frequencies. At high frequencies the frequency independent and slow temperature dependent variation is attributed to electronic and ionic polarisation [20]. The absence of peaks in the loss (tanδ) versus temperature curves indicate the absence of dipolar impurities.The value of ε^{11}at low frequencies is found to increase to very high value due to free charge motion within the material and is connected to AC conductivity relaxation [21]. The strong dispersion in both the components ε^1 and $\varepsilon^{11}$of the complex dielectric constant appears to be common feature in rocks and is referred to as low frequency dispersion [15].

It is further observed that there is a strong dispersion of ε^1, tanδand ε^{11}at low frequencies and high temperatures and small dispersion at high frequencies and low temperatures in case of samples (a) and (b); whereas the dispersion is relatively very low as function of frequency and temperature in case of sample (c). The enhanced dielectric behaviour in samples (a) and (b) in comparison with sample (c) is attributed to the presence of relatively higher percentageof Fe_2O_3 componentin the granite composition. On the whole the variation of ε^1, tanδand ε^{11}follow the usual trend of dielectric behaviour[15].

D. Electric modulus

The dielectric response of the materials can alternately be analysed in terms of complex electric modulus M*(ω), with added advantage of suppressing the electrode polarisation effects.Physically, the electric modulus corresponds to the relaxation of the electric field in the material when the electric displacement remain constant, so that the electric modulus represents the real dielectric relaxation process, which can be expressed as[22],

$$M^*(\omega) = \frac{1}{\varepsilon^*(\omega)} = M^1 + iM^{11} = M_\infty \left[1 - \int_0^\infty \left(-\frac{d\phi(t)}{dt} \right) \exp(-i\omega t)\, dt \right] \qquad (1)$$

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Where $M^1(\omega)$ and $M^{11}(\omega)$ are real and imaginary parts of electric modulus; $M_\infty=(\varepsilon_\infty)^{-1}$ is the asymptotic value of $M^1(\omega)$ and $\emptyset(t)$ is the electric field within the material.

The representative of curves of the $M^1(\omega)$ and $M^{11}\omega)$ of the electric modulus at different temperatures for granite samples are shown in fig.5.

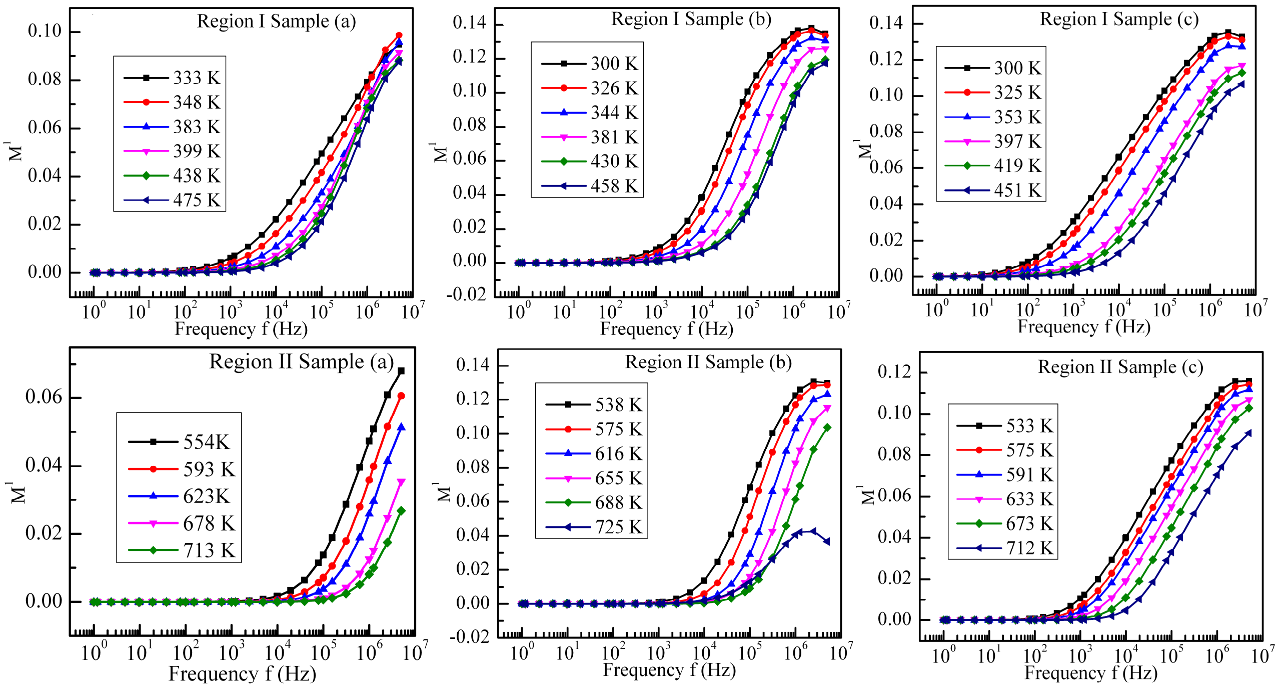

Fig. 5.M^1 as a function of frequency of samples (a), (b) and (c) for two temperature regions

It can be seen from the figures that $M^1(\omega)$ attains almost a constant value at high frequencies for all temperatures except for sample (a) and tends to zero at low frequencies, indicating negligible electrode polarisation. In the case of sample (a),the dispersion is observed even at high frequencies due to presence of relatively higher content of Fe_2O_3 in the sample.

The $M^{11}(\omega)$ spectra in fig.6 show slightly asymmetric peaks which shifts towards higher frequencies as temperature increases.

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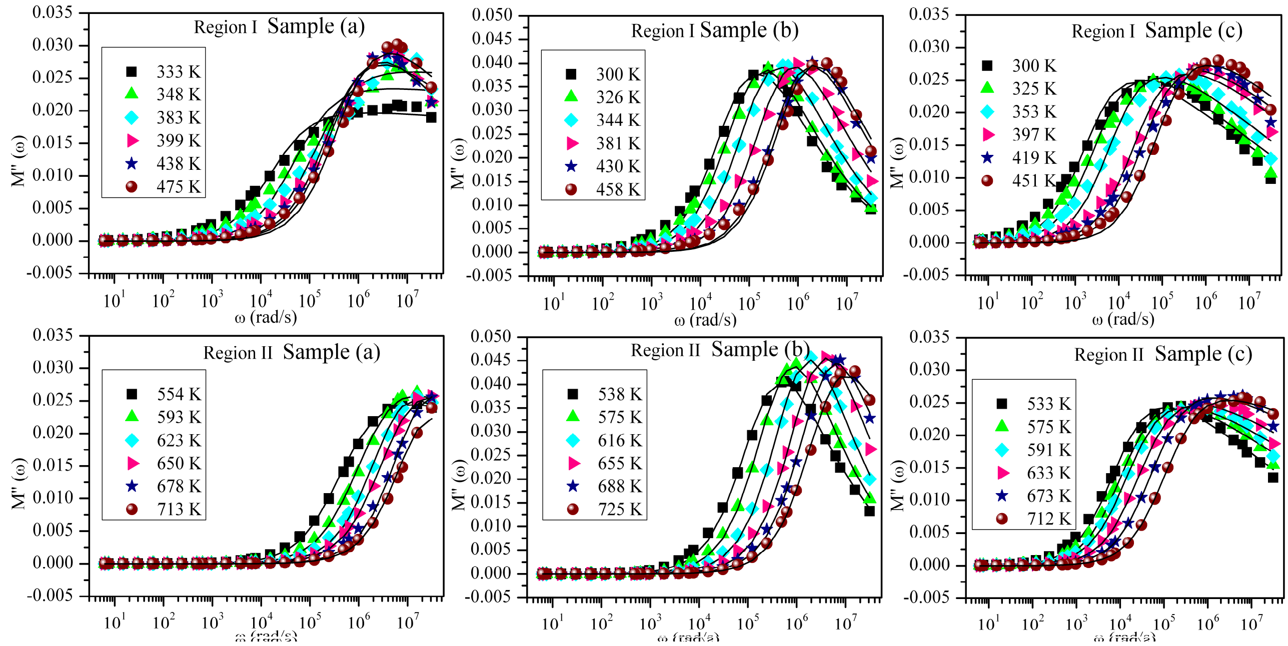

Fig. 6. M^{11}as a function of frequency of samples (a), (b) and (c) for two temperature regions and the solid lines of the plots are Bergman equation fits.

These peaks indicate the transition from short range to long range mobility with decreasing frequency, where the low frequency side of the peak represents the range of frequencies in which the ions are capable of moving long distances, i.e. performingsuccessful hoping from one site to the neighbouring site,whereas for the high frequency side, the ions are spatially confined to their potential wells and can execute only localized motion [23].

The dielectric relaxation process, in general can be represented by the numerical Laplace transform of theKohlrausch-William-Watts (KWW) decay function,

$$\phi(t) = \exp\left(-\frac{t}{\tau}\right)^{\beta} \qquad (2)$$

Here the exponent β characterizes the degree of non-Debye behaviour and τ is the conductivity relaxation time [25].

Bergman [24] gave the modified KWW function fitting approach which allows direct analysis in the frequency domain[25]. The imaginary part $M^{11}(\omega)$ has been approximated as (for $\beta \geq 0.4$);

$$M^{11}(\omega) = \frac{M^{11}_{\max}}{(1-\beta)+\left(\frac{\beta}{1+\beta}\right)\left[\beta\left(\frac{\omega_{\max}}{\omega}\right)+\left(\frac{\omega}{\omega_{\max}}\right)^{\beta}\right]} \qquad (3)$$

Where $M^{11}_{\max}$ is the peak maximum of the imaginary part of the modulus and $\omega_{\max}$ (=1/τ) is the peak frequency of the imaginary part of the modulus. We analysed our experimental data using this Bergman approach. It is clear from fig.6 that the Bergman frequency domain function fits with our experimental data both at low and high frequencies.It is predicted thatthe β values are greater than 0.4 indicating correlated motions between the ions;further the nature of relaxation is attributed to be non-Debye type.

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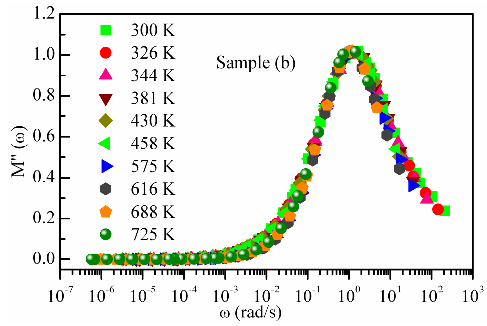

Fig.7. Typical plots of normalized M^{11} versus ω for the sample (b) at various temperatures

Fig. 7 shows the typical plot of normalized imaginary part of the modulus versus normalized ω for granite sample (b). The data for different temperatures are found to overlap almost perfectly except on the high frequency tail. This indicates clearly that all dynamic processes occurring at different time scales exhibit the same activation times is independent of temperature.

Following the relation:

$$\omega_{max} = \omega_0 \exp(-E_a/k_B T) \qquad (4)$$

The activation energies are evaluated from the curves of $M^{11}(\omega)$ versus frequency at different temperatures. The average activation energies obtained in two temperature regions are 0.34eV (region I) and 0.51eV (regionII).

IV. CONCLUSIONS

In the present investigation the XRF studies indicated that the major components of the granite samples are SiO_2,Al_2O_3 and Fe_2O_3. FESEM studies indicated that larger grain size with more crystallinity. The enhanced dielectric behaviour in the samples (a) and (b) is attributed to relatively higher content of Fe_2O_3. The absence of peaks in dielectric loss and Bergman curve fitting approach are indicative of non-Debye type relaxation.The perfect overlap of the normalized modulus spectra obtained for different temperatures indicates that the distribution of relaxation times is independent of temperature.

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