JOURNAL OF MATERIALS SCIENCE **40** (2005) 4435–4437 LETTERS

Categorizing dielectric dispersion using the multiple-arc approach

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Valuable experimental data have been published in recent years concerning the anomalous behavior of dielectric materials with frequency [1–5]. Improvement and development of engineering applications of these materials is often hindered by the lack of a comprehensive understanding of the mechanisms underlining the variation of dielectric parameters with frequency of the applied field, the sintering or forming temperatures used in the production of the material as well as the temperatures encountered in operation. The anomalous behavior of dielectric material is manifested by frequency dispersion corresponding to wide range of relaxation [6–8]. Significant advances have been made regarding the physical interpretation of the behavior of the various dielectric parameters on both microscopic and macroscopic scales [9–15]. However, practical applications of dielectric materials demand the development of suitable tools that are capable of modeling the behavior exhibited by these materials so that corrective procedures can be devised to alter or modify the prevailing characteristics to those more suited to specific applications. The aim of the present letter is to report an approach to categorize dispersion in dielectric materials. This approach is based on multiple-arc analysis [14]. In this method, the relaxation time distribution (RTD) takes the form

$$f(u) = C \sum_{n=1}^{m} \frac{(\varepsilon_{sn} - \varepsilon_{\infty n}) \sin(\alpha_n \pi)}{\mathrm{Cosh}[(1-\alpha_n)(u+\beta_n)] - \cos(\alpha_n \pi)} \tag{1}$$

where $C = [2\pi \sum (\varepsilon_{sn} - \varepsilon_{\infty n})]^{-1}$, $\beta_n = \ln(\tau_{01}/\tau_{on})$, $u = \ln(\tau/\tau_{01})$, with $\int_{-\infty}^{\infty} f(u)\mathrm{d}u = 1$. ε_{sn} and $\varepsilon_{\infty n}$ are the low- and high-frequency dielectric constants of the nth arc, respectively, while m designates the number of arcs traced over the frequency range in use with α_n as the spread parameter for the nth arc taking values between 0 and 1. τ_{on} represents the most probable relaxation time for the nth arc, while τ_{o1} represents an arbitrary chosen low frequency reference arc. Each term in the above distribution corresponds to a single-arc distribution multiplied by the weighting factor,

$$(\varepsilon_{sn} - \varepsilon_{\infty n}) \left[\sum_{n=1}^{m} (\varepsilon_{sn} - \varepsilon_{\infty n}) \right]^{-1}$$

It is worth pointing out that the above distribution resembles a statistical average formula for different single-arc distributions, presumably reflecting various categories of polarization and phase transition modes occurring in the materials subjected to varying frequency and temperature [16]. In multiple-arc analysis, the real and imaginary components of the complex dielectric constants $\varepsilon^* = \varepsilon' - \mathrm{j}\varepsilon''$ are expressed as [14],

$$\varepsilon'(\omega) = \varepsilon_{\infty n} + \sum_{n=1}^{m} (\varepsilon_{sn} - \varepsilon_{\infty n}) \int_{-\infty}^{\infty} \frac{f(u)}{1+\omega^2\tau^2} \mathrm{d}u \tag{2}$$

and

$$\varepsilon''(\omega) = \sum_{n=1}^{m} (\varepsilon_{sn} - \varepsilon_{\infty n}) \int_{-\infty}^{\infty} \frac{\tau\omega f(u)}{1+\omega^2\tau^2} \mathrm{d}u \tag{3}$$

where ω is the angular frequency of the applied field. A detailed account of data analysis using the multiple-arc approach is reported elsewhere [14]. The data treated here are those reported by Sundrakaurma *et al.* [15], which comprises dielectric and conductivity measured results for potassium niobate ($KNbO_3$) ferroelectric ceramics as a function of temperature, T, (100 to 200 °C) and frequency, f, (10^2 to 10^6 Hz). Their analysis, and due interpretations, made use of the Cole–Cole functions in terms of $\varepsilon'(\omega)$and $\varepsilon''(\omega)$. As indicated in their paper, the applicability of these functions is rather limited to the lower frequency range of measurements, thereby leaving the high frequency range of data unattainable by the Cole–Cole analysis. In our approach, presented here, the multiple-arc method is employed to analyze the reported measured data. Following the procedure outlined in [14], two arcs are obtained covering the whole range of frequency used. Only two sets of results are presented here corresponding to high temperature, 197 °C, and low temperature, 116 °C, and their arc parameters are given in Table I.

Using Equations 2 and 3 with the parameters in Table I, remarkable agreements are realized between computed and measured data in terms of $\varepsilon'(\omega) - \varepsilon''(\omega)$ relations as shown in Fig. 1. The high frequency data generate new arcs in addition to the low frequency arcs reported earlier [15]. However, apparent agreement in $\varepsilon'(\omega) - \varepsilon''(\omega)$ relation does not imply validation of the new approach. Therefore, the compliance of data with the analysis is further examined through $\varepsilon' - \omega$ and $\varepsilon'' - \omega$ relations. Fig. 2 depicts satisfactory agreements

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TABLE I Dielectric dispersion parameters for KNbO3 ceramics

	Arc I (low frequency)				Arc II (high frequency)			
T (°C)	ε_s	ε_∞	α	$\tau_o\,(\mu s)$	ε_s	ε_∞	α	$\tau_o\,(\mu s)$
116	1070	755	0.6	5500	765	565	0.32	0.025
197	1385	995	0.53	165	995	772	0.1	0.03

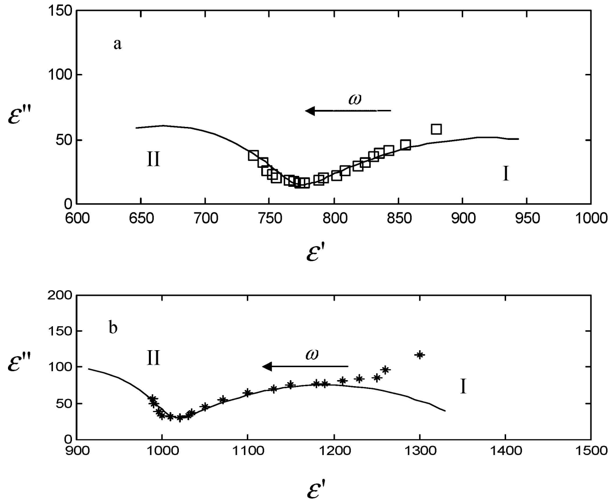

Figure 1 Cole–Cole plot for KNbO3 ceramics. □, *: measured, ——: computed (a) $T = 116\,°C$, (b) $T = 197\,°C$.

between measured data and computed dielectric constant particularly in the high frequency parts, which are missing in the Cole–Cole analysis reported in [15]. However, there are deviations in the computed results at the high temperature and low frequency range. This is likely to be attributed to losses caused by the onset of dc conduction rather than dielectric polarization mechanism, thus the dispersion of dielectric constant does not comply with arc analysis. Using the parameters in Table I and Equation 1, the relaxation time distributions are then obtained for both temperatures as depicted in Fig. 3. Both distributions feature two peaks corresponding to the low- and high-frequency arcs. The low frequency arcs are thoroughly discussed in [15], and ascribed mainly to the decay of polarization due to oxygen defect-related dipoles. In our study, the following features can be extracted from the distributions.

(i) There are no significant changes in the relaxation magnitude $\Delta\varepsilon$ $(\varepsilon_s - \varepsilon_\infty)$ for both arcs. The relative small differences $\Delta\varepsilon$ may be regarded as within small error in fitting the arcs.

(ii) The spread parameter, α, is not altered substantially with temperature for the low frequency arc while it increased markedly at lower temperature for the high frequency arc. This is reflected in the sharpness of the RTD at high temperature.

(iii) The characteristic time τ_o is reduced with the increase in temperature for low frequency arc while it is increased insignificantly with temperature for the high frequency arc.

(iv) There is little or no overlap of arcs in the data for both temperatures. This may suggest that two different categorizes of polarization are taking place.

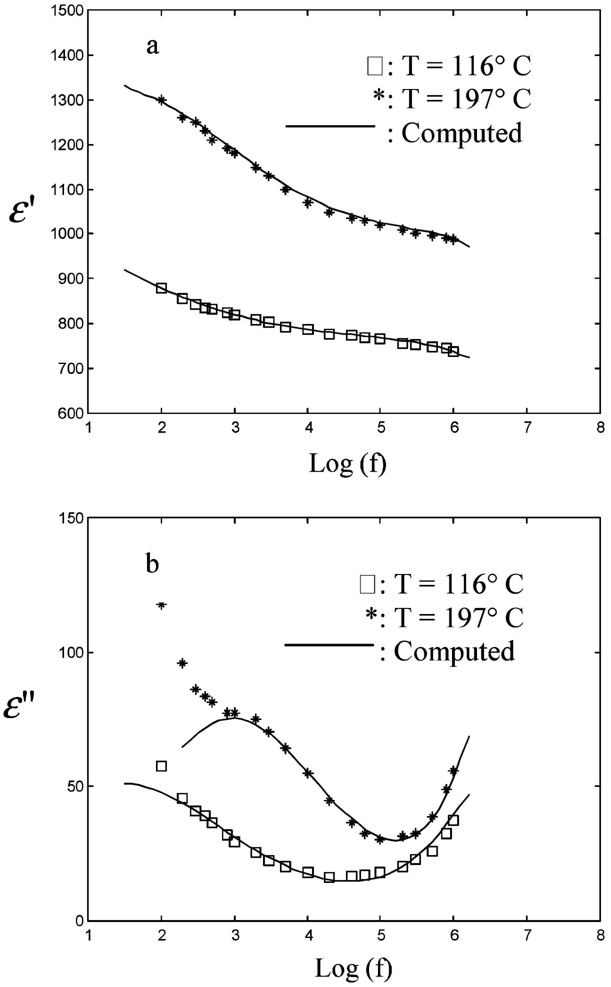

Figure 2 Comparison between computed and measured dielectric constant of KNbO3 ceramics. (a) Real component (b) Imaginary component.

Direct inspection of Table I indicates that two distinctly different polarization mechanisms characterizes the dispersion of KNbO3 ceramics. As mentioned above, the low-frequency KNbO3 has been thoroughly investigated in [15] and subsequently attributed to defect-related relaxation whereby the lack of KNbO3 stoichiometry results in oxygen vacancy dipoles. Using Arrhenius equation, $\tau = \tau_o' \exp(E_R/k_B T)$, and the low-frequency relaxation times, from Table I, the activation energy E_R of relaxation process is evaluated to be 0.691 eV, and the pre-exponential factor τ_o' to be 7.2×10^{-12} s. Both values are in reasonable agreement with those reported in [15].

However, different trends in parameters are realized for the high frequency arcs indicating a different mechanism of polarization. Here, attributes of KNbO3 ferroelectricity are considered where the increase of temperature causes further randomization of the permanent dipoles weakening their effects [17]. This in turn reduces the mutual interactions between dipoles which are regarded as the prime cause for the spread in relaxation [18]. That may explain the reduction in the spread parameter,α, for high temperature as indicated in Fig. 3. It is also evident from Fig. 2, that the increase in temperature enhances polarization and increases losses in the material for both arcs. The overall trend of the reported data remains almost the same over the whole

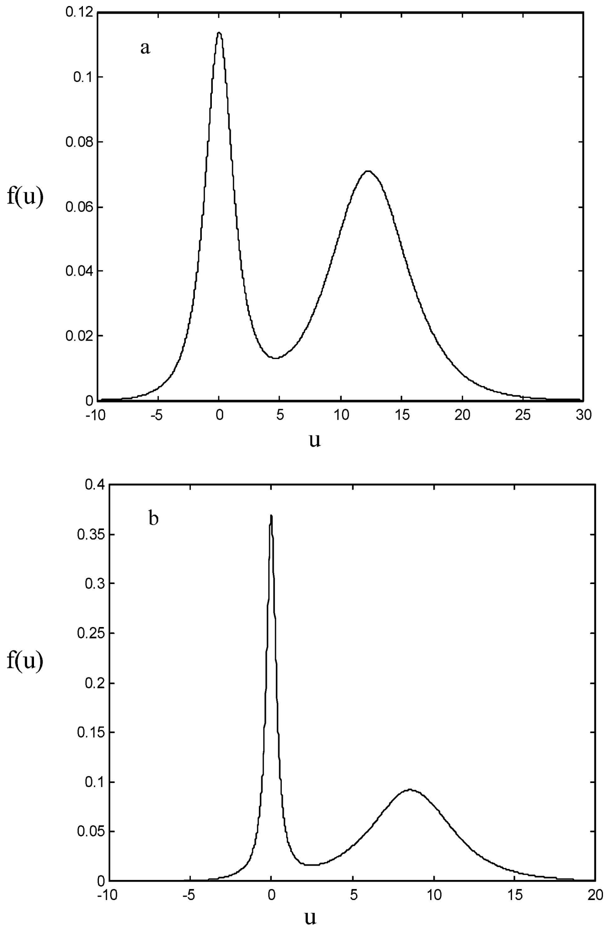

Figure 3 Relaxation time distribution function for $KNbO_3$ ceramics. (a) $T = 116\,^{\circ}C$, (b) $T = 197\,^{\circ}C$.

temperature range used [15]. Thus, it is expected that the remaining data on $KNbO_3$ ceramics within this temperature range are characterized by features similar to those reported here, and in this respect further work is underway and will be reported in due course.

In so far, the use of multiple-arc approach has revealed further features in $KNbO_3$ ceramics dispersion. The capability of the analysis in covering the whole range of frequency used has suggested a new polarization mechanism at high frequency, which was unattainable by the well-known Cole–Cole single-arc analysis. It is anticipated that this work can be further developed to cover a wide range of materials and operating conditions, and categorize the dielectric polarization and relaxation mechanisms, so that existing practical applications can be improved and new applications be explored.

References

1. C. ANG, R. GUO, A. S. BHALLA and L. E. CROSS, *J. Appl. Phys*. **8**(10) (2000) 7452.
2. J. C. BADOT, V. BIANCHI, N. BAFFIER and N. BELHADJ-TAHAR, *J. Phys.: Condens. Matter*, **14** (2002) 6917.
3. C. ANG, Z. YU, H. J. YOUN, C. A RANDALL, A. S. BHALLA, L. E. CROSS, J. NINO and M. LANAGAN, *Appl. Phys. Lett.* **80**(25) (2002) 4807.
4. V. KAPUSTIANIK, M. FALLY, H. KABELKA and H. WARHANEK, *J. Phys.: Condens. Matter* **9** (1997) 723.
5. D. K. GOEL, C. P. SINGH, R. K. AHUKLA and A. KUMAR, *J. Mater. Sci.* **35**(4) (2000) 1017.
6. H. FAN, L. ZHANGAND and X. YAI, *ibid.* **34**(4) (1998) 895.
7. D. K. DURGA and N. VEERAIAH, *ibid.* **36**(23) (2001) 5625.
8. L. K. PAN, H. T. HUANG and C. Q. SUN, *J. Appl. Phys.* **94** (2003) 2695.
9. E. NEAGU, P. PISSIS, L. APEKIS and J. L. GOMEZ RIBELLES, *J. Phys. D: Appl. Phys.* **30** (1997) 1551.
10. S. N. AL-REFAIE, *J. Mater. Sci.* **32**(23) (1997) 6221.
11. E. TUNCER, S. M. GUBANSKI and B. NETTELBLAD, *J. Appl. Phys.* **89**(12) (2001) 8092.
12. V. BANERJEE and S. DATTAGUPTA, *Phys. Rev. B* **68** (2003) 054202(8).
13. D. DEGER and K. ULUTAS, *J. Appl. Phys.* **89**(12) (2001) 8101.
14. H. S. B. ELAYYAN and S. N. AL-REFAIE, *IEE Proc. Sci. Meas. Technol.* **150**(4) (2003) 141.
15. B. SUNDARAKANNAN, K. KAKIMOTO and H. OHSATO, *J. Appl. Phys.* **94**(8) (2003) 5182.
16. S. N. AL-REFAIE, *Semicond. Sci. Technol.* **19** (2004) 208.
17. V. V. DANIEL, 'Dielectric relaxation' (Academic Press, New York, 1967) Chapter 16.
18. *Idem.*, 'Dielectric relaxation' (Academic Press, New York, 1967) Chapter 8.

Received 9 September 2004
and accepted 18 January 2005