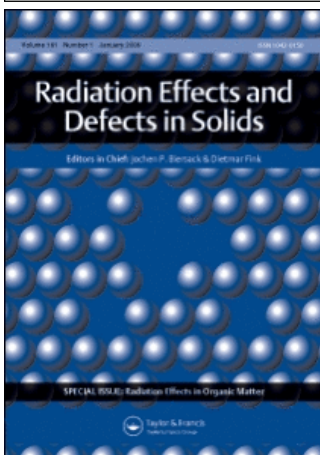


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Thermal depolarization spectroscopy for probing the contribution of CaCO_3 to the dielectric relaxation of dolomite ($\text{CaMg}(\text{CO}_3)_2$)

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THERMAL DEPolarIZATION SPECTROSCOPY FOR PROBING THE CONTRIBUTION OF CaCO_3 TO THE DIELECTRIC RELAXATION OF DOLOMITE ($\text{CaMg}(\text{CO}_3)_2$)

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The Ionic Thermocurrent (ITC) technique probes a relaxation mechanism in both polycrystalline and monocrystal dolomite, at 189 K. Dielectric characterization shows that it is dipolar and is probably related to the defect structure of the calcium sublattice.

1 INTRODUCTION

The knowledge of the physical properties of carbonates can extend their industrial applications and uses.¹ Dolomite ($\text{CaMg}(\text{CO}_3)_2$) is a mixed ionic material proper for studying the dielectric relaxation in relation to the end members relaxation. In the present work, the Thermal Depolarization (TD) or Ionic Thermocurrent technique (ITC) is used as a probing method for deeper insight of ionic relaxation.

2 EXPERIMENTAL DETAILS

Polycrystalline dolomite from Greece and monocrystals from Oberdorf, Austria were studied. The chemical analysis results for the polycrystals are: Loss on ignition: 46.91%, SiO_2 :0.20%, Al_2O_3 :0.12%, Fe_2O_3 :0.04, CaO :30.51%, MgO :21.81%. Information about the monocrystals have previously been reported.² Additional atomic absorption analysis for the impurity content of polycrystals gave: Mn^{2+} :0.02%wt, Sr^{2+} :0.02%wt and for the monocrystals Sr^{2+} :0.02%wt. Details about the ITC apparatus used has been given elsewhere.³

3 RESULTS AND DISCUSSION

An intense peak (Figure 1a) of the low temperature thermal depolarization spectra of polycrystal dolomite maximizes at $T_M = 189$ K. The peak positioned around 140 K is related to the Mg^{2+} sublattice, because it is also detected in MgCO_3 (the results will appear in a forthcoming paper) while it is completely absent in CaCO_3 .⁴ The 189 K band is insensitive to the electrode material used (platinum, bronze, teflon, silver paste, graphite) and to the sample thickness, by keeping E_P the same. The results strongly indicate the dipolar character of the mechanism. Annealing at 400°C for 30 min and subsequent quench to room temperature results in a decrease of the peak amplitude (Figure 1a). The mechanism is also insensitive to mechanical treatment (bending) of the sample. Peak cleaning³ isolates the dominant mechanism with parameters: $E = 0.53$ eV,

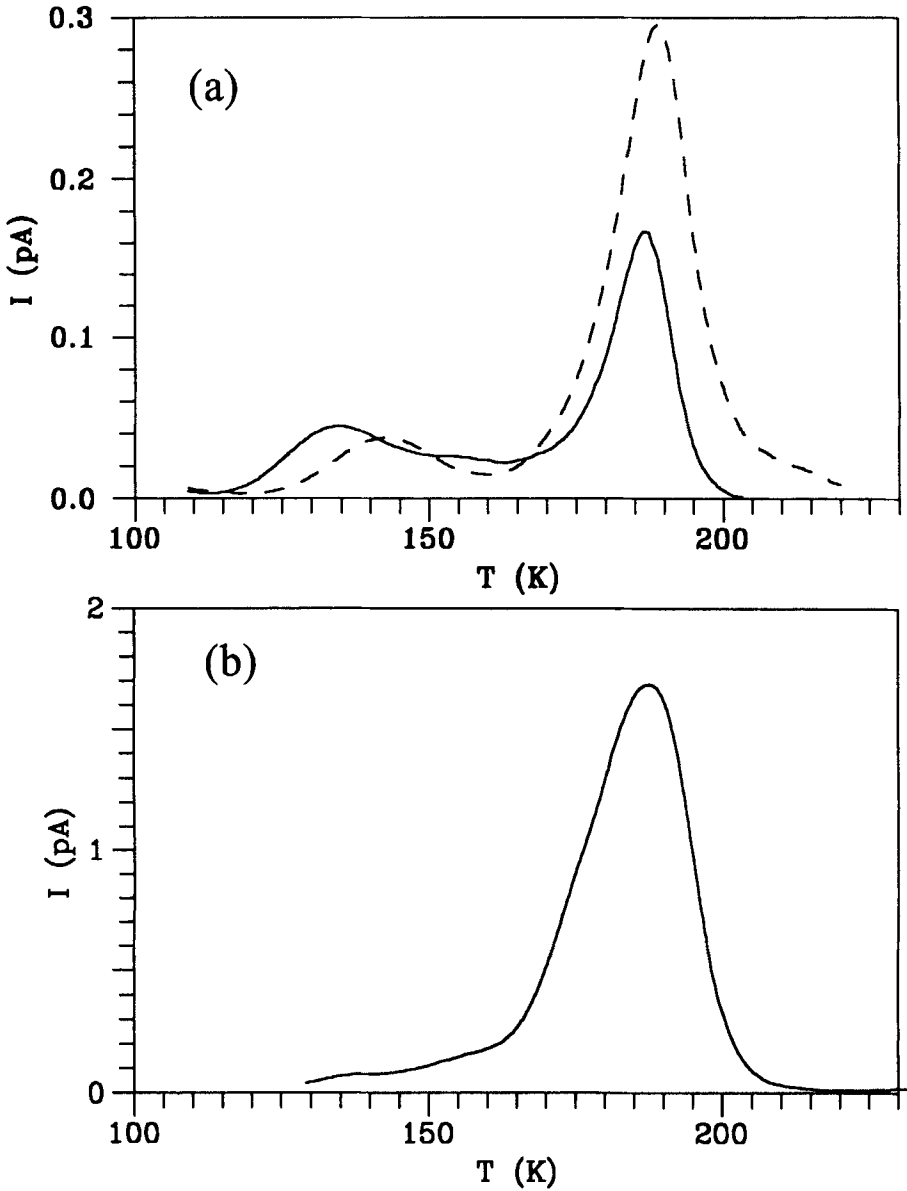


FIGURE 1(a) ITC of as received (dashed line) and thermally treated at 400°C for 30 min (solid line) polycrystalline dolomite. In both cases the sample was polarized at $T_p = 200\text{ K}$. The heating rate was 3 K/min . (b) Isolation of the 189 K band in monocrystal dolomite by polarizing at $T_p = 180\text{ K}$ for a too short period of time ($t_p = 1-2\text{ sec}$).

$\tau_0 = 1.22 \times 10^{-11}$ sec. By partial heating³ we get $E = 0.5-0.62$ eV. In monocrystal dolomite we have also isolated a peak located at $T_M = 189$ K as well (Figure 1b). Due to neighbourhooding overlap, we evaluated by partial heating³ the activation energy to be $E = 0.5-0.75$ eV. The above mentioned dispersion is also detected in calcite (CaCO_3)⁴ while, in our preliminary experiments, is absent in MgCO_3 . Consequently, it involves the relaxation of the defect structure of the calcium sublattice. The dipolar characteristics and the thermal treatment probably indicate the existence of impurity-vacancy complexes. By comparison of the impurity content of all carbonates studies we speculate that the defect dipoles come from Ca^{2+} substitution by Al^{3+} . The distribution of Sr^{2+} to interstitial⁵ or Ca^{2+} lattice sites in relation to the lattice constant of carbonates may also favour the creation of defect dipoles. A previous⁴ explanation for the rotation of distorted CO_3^{2-} tetrahedra is rejected as it should stimulate the same band also in MgCO_3 .

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