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INHOMOGENEOUS STRAIN INDUCED BY FAST NEUTRON IRRADIATION IN $NaKSO_4$ CRYSTALS *

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ABSTRACT

The effect of fast neutron irradiation on the thermal properties of $NaKSO_4$ crystals was studied around the phase transition temperature $T_c = 453$ K. The thermal expansion coefficient as well as the phase transition temperature were found to be dependent upon the irradiation dose. The specific heat, C_p , showed multiple peaks in the phase transition temperature region.

An explanation of this behaviour was based on the induced inhomogeneous strain in the crystal caused by the neutron irradiation process.

INTRODUCTION

A characteristic first order phase transition of a solid-solid transformation in $NaKSO_4$ crystals was observed using thermal analysis, infrared and x-ray diffraction techniques (1).

Previous studies of thermal properties of $NaKSO_4$ crystals showed an anomaly in the temperature dependence of specific heat (Cp), linear thermal expansion coefficient α_{ij} and heat transition ΔQ at 453 K (2). These observations were confirmed by the results of the electrical conductivity in the same temperature range (3).

$NaKSO_4$ crystals irradiated by fast neutrons of energy 14.5 Mev. The electrical conductivity measurements in the temperature range 300-500 K showed a pronounced change in the phase transition temperature (4).

In this paper, the effect of neutron irradiation on the thermal properties of $NaKSO_4$ crystals, in the same temperature range, was studied. The results were discussed in terms of the induced in homogeneous strain.

EXPERIMENTAL

a. Crystal preparation

Stoichiometric ratios of Na_2SO_4 and $K_2 SO_4$ were added to produce $NaKSO_4$ crystals which were purified five times. Large single crystals were grown up by means of slow dynamical isothermal evaporation methods (5).

Chemical analysis, x-ray diffraction and optical inspection were used for purity control and to insure the quality of the crystal.

b. Thermal measurements

A Heraeus TMA 500 dilatometer was used for the thermomechanical analysis measurements. The sample temperature was monitored by means of a Ni-Cr-Ni thermocouple fitted in the sample holder of the standard design which was made of quartz. The linear expansion coefficient was calculated on the assumption that the quartz expansion coefficient has insignificant effect on the measured values, the heating rate used was 2K min^{-1}. The samples were shaped in the form of rectangular rods 2X2X20 mm^3 out from a large single crystal of $NaKSO_4$. The longer part of the sample was oriented along the crystallographic direction (001).

The specific heat under constant pressure, C_p was determined using a differential scanning calorimeter (DSC) technique, where a Heraeus DSC cell was connected to the Heraeus DTA 500 thermal analyser. Measurements were performed by applying the base line methods (6). Lidded pans, made of aluminium, were used to eliminate the sloping of the base line. A Pt-100 thermocouple was used as temperature sensor, while a heating of 2k min^{-1} was applied.

c. Irradiation process

The samples were irradiated by different neutron doses at room temperature (298 K) using a compact D-T neutron generator (Phillips product Pw 5310) of known neutron spectrum (4).

The neutron flux in each run was controlled using Cu foil as a monitor through ^{63}Cu (n, 2n) ^{62}Cu reaction. A neutron beam of energy 14.5 MeV and flux $4X10^5$ n cm^{-2} sec^{-1} was used up to 12 hours of irradiation. Radioactivities within the sample were determined using G.M. detector and scintillation NaI detector (4).

RESULTS AND DISCUSSION

Non irradiated $NaKSO_4$ crystals, when heated, showed an anomaly at 453 K (2), where the thermal expansion coefficient measured along the Z axis had a negative value.

The samples showed different thermal behaviour when irradiated with different neutrons doses ranging from 0.9 up to 4.8 Rad.

The extent of the negative part of the dilatometric curve varied with different doses. In addition, the temperature at which the anomaly occured has changed with the irradiation dose.

Figure (1) showed that in the temperature range 340-470 K, where the crystal shows anomalous behaviour, $\Delta L/L_o$ tends to have a negative value. The maximum anomalous shrinkage occured when the crystal was irradiated by 1.9 Rad neutron dose. However, the degree of shrinking

was less when the irradiation dose has deviated from this value. This behaviour can be attributed to the decomposition induced by irradiation as well as the generation of new species (7).

The decomposition products may have been trapped in the interstitial positions of the crystal lattice producing stresses which clamp the domains and cause the anomaly in the crystal properties.

The various values of the spontaneous tensile strain η_{33} were calculated using the method described by previous workers (8,9). The results are represented in figure (2 a).

The variation of spontaneous strain at T_c with neutron irradiation dose was presented in Fig. 2.b. The results can be interpretted by assuming that two processes have taken place in the crystal lattice, namely the release of new species as well as the trapping process. Both processes depend on the dose of neutron irradiation. At small doses the trapping process was assumed to be negligible and the increased concentration of generated species played the dominant role. This may explain the generation of the high strain induced region (AB) inside the crystal. These generated species needed more thermal energy to enhance their mobility.

As the irradiation dose increases the activity of the trapping centres becomes the dominant factor leading to partial release of such induced stress (10).

During our previous studies of the thermal properties of $NaKSO_4$

crystals, anomalous behaviour of the specific heat (C_p) was observed as a single peak at 453 K (2). Upon irradiation of the crystal the position of the peak representing the change in C_p with temperature, was found to vary with the irradiation dose. The lowest specific heat value was obtained at a neutron dose ≈ 2.1 Rad. This is in agreement with the thermomechanical measurements which showed maximum stress at the same irradiation dose (10). Figure (3) also shows that a multiple peak pattern of the C_p change, appeared at a higher dose. At 5.3 Rad two distinct peaks were clearly identified and were attributed to inhomogeneous internal strain generated by irradiation.

Finally, the presence of these multiple peaks excludes the possibilities that they were due to sample handling or crystal growth conditions.

In conclusion, the induced inhomogeneous strain proved to be sensitive to the neutron irradiation dose and could thus be utilized in detecting the neutron exposure.

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REFERENCES

1. M.S. Kumair and E.A. Secco, Can. J. Chem. 61, 594 (1983).
2. M. E. Kassem, S. H. Kandil, E. F. El-Wahidy and M. El-Gamal, Revue phys. Appl. 19, 445 (1985).
3. M. E. Kassem, E. F. El-Wahidy, S. H. Kandil and H. Gado, Ferroelectrics letters 5, 71 (1986).
4. M. E. Kassem, A. M. El-Khatib, E. F. El-Wahiby and H. E. Gado, J. Mat. Sci. Lett. accepted.
5. B. M. Bartlett, J. Sci. Instr. 38, 54 (1961).
6. T. Daniels in "Thermal Analysis" Kogan Itd. London P. 127 (1973).
7. K. Okada, J. A. Gonzalo and J. M. Rivera J. Phys. Chem. Solids GB 28, 689 (1957).
8. S. Shiozaki, A. Sawada, Y. Ishibashi and Y. Takagi J. Phys. Soc. Japan, 42, 353 (1977).
9. T. Oska, J. Phys. Soc. Japan 45, 571 (1978).
10. G. M. Laiacone, M. Delfino, A. Shaulov, W. A. Smith and M. I. Bell, Ferroelectrics 29, 181 (1980).

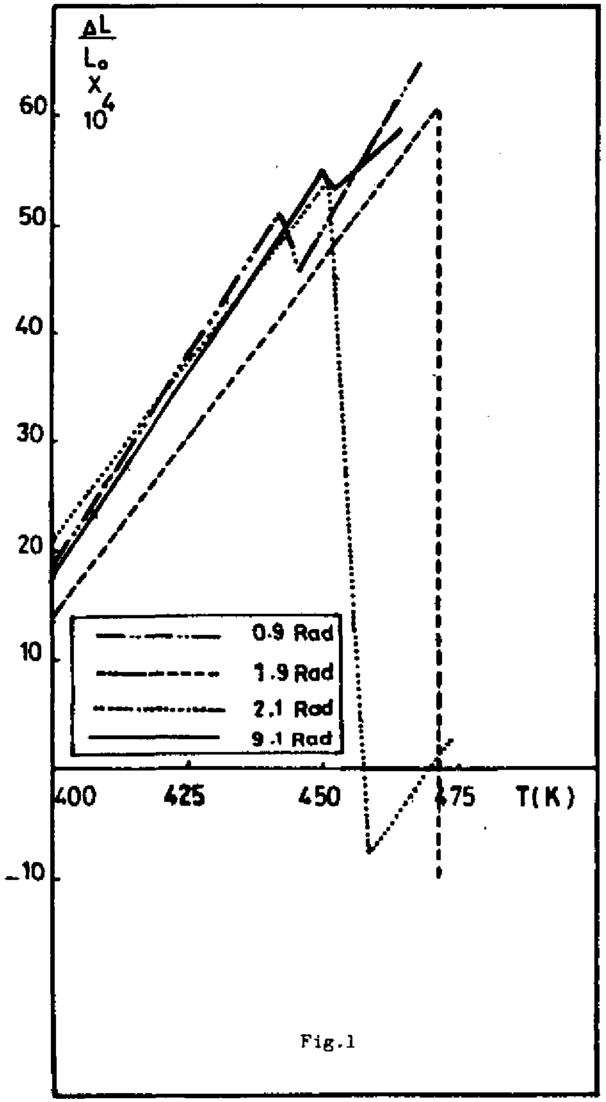
ΔL / L₀ X 10⁴
60
50
40
30
20
10
-10
400
425
450
475
T(K)
0.9 Rad
1.9 Rad
2.1 Rad
9.1 Rad

Fig.1

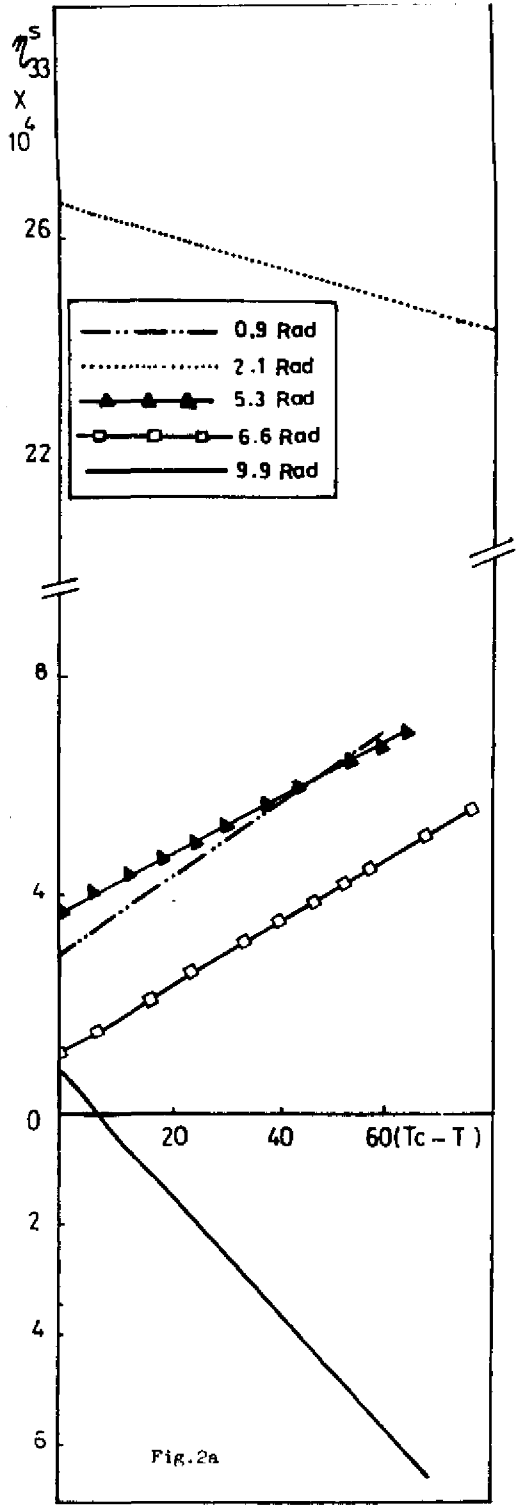
η^s_33 X 10⁴
26
22
8
4
0
2
4
6
20
40
60(Tc – T)
0.9 Rad
2.1 Rad
5.3 Rad
6.6 Rad
9.9 Rad

Fig.2a

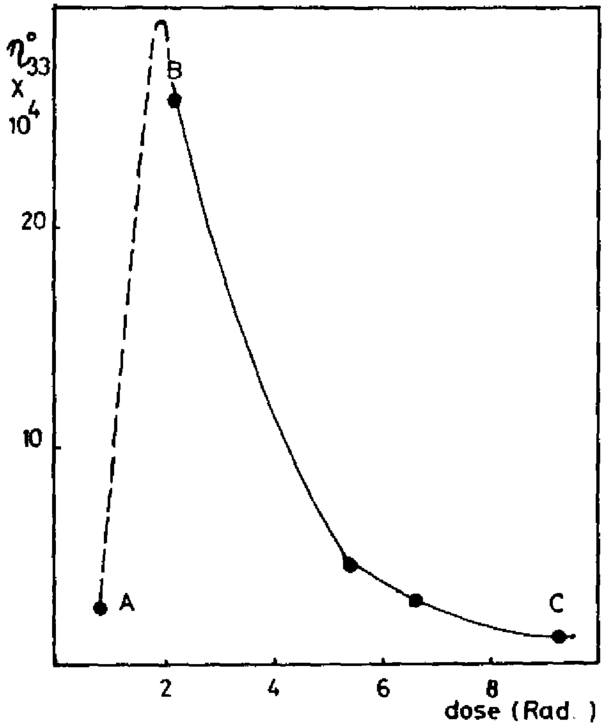
η°33 x 10⁴
20
10
A
B
C
2
4
6
8
dose (Rad.)

Fig.2b

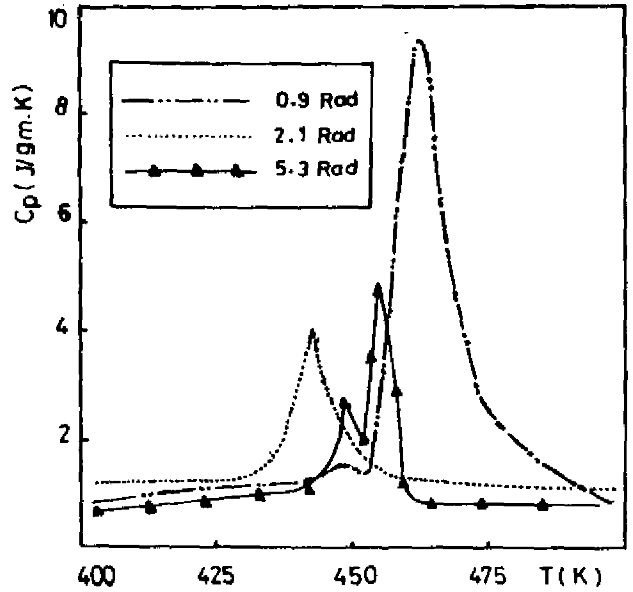
Cp (J/gm.K)
10
8
6
4
2
0.9 Rad
2.1 Rad
5.3 Rad
400
425
450
475
T (K)

Fig.3