Physical Radiation Processes in the Solid State Dielectrics

I.Kh. Abdukadirova

Institute of Nuclear Physics Academy of Sciences Uzbekistan
702132 Tashkent. Uzbekistan. Ulugbek

Abstract – The process of radiation damage and phase transition in solid state dielectric – alpha-quartz has been determined by spectroscopic methods – Raman light scattering and infrared spectroscopy. The influence of radiation on spectral characteristics of the quartz crystal lattice of main vibrations was established in region of a frequencies 100–1200 cm^{-1} in Raman and infrared spectra. At the intensive phase transition was discovered in the spectrum transformation in the region of valent and compound vibrations. A kinetics change of radiation effects for spectral characteristics of the "soft" modes is given. The problems connected with the possiblity of mechanism radiation-induced phase transformation in quartz were discussed. The effect of radiation on the photorefractive, permitivity and polarizability advanced solid state dielectric materials: crystalline quartz, quartz glass irradiated with different neutron fluxes were adequately.studied

1. Introduction

The question about of radiation spectroscopy of different modifications of silica is studied insufficiently and it is necessary to develop this investigations. In most cases we examined the influence on luminescence of silica crystal or glass [1–7]. Authors [2,7] usually using small irradiation doses, different samples and different irradiation sources, mostly applying gamma-irradiation and ion implantation [4,5].

More often it concerns to learning of radiation spectroscopy questions with wide using in science and techniques alpha-quartz modification [1,5]. In particular there is no exhaustive information [4,7] about problem of neutron-activated phase transition in the quartz by method of oscillation spectroscopy, when the silica is subjected to long time exposition in nuclear reactors channel.

In this work one crystalline modification: alpha-quartz was studied. The plates with identical history, purity and geometry are investigated. The samples of the crystalline quartz were exposed with different fast neutrons fluxes in channels of a WWR-SM reactor.

As a result of high fluxes of fast neutrons bombardment the single crystals of alpha-phase are transformed into another crystalline phase. This process of radiation damage of crystalline structure and defect formation has been studied by spectroscopic methods: Raman light scattering and infrared spectroscopy for receiving the information about a kinetics of process, about a volume effect of radiation spectroscopy. The Raman spectra and the infrared spectra were measured before and after irradiation with the fluxes (F) of a samples.

The effect of radiation on the photorefractive, permitivity and polarizability advanced oxides materials have been adequately studied. In this work of two neutron – irradiated modifications of the fused quartz: low-temperature of crystalline quartz(I) and quartz glass (II) were studied. We investigated the plates with some history, pure and geometry. The samples I-II were irradiated with different neutron fluxes in the channels of a WWR-SM reactor.

2. Results and discussion

The influence of radiation on spectral characteristics of the quartz crystalline lattice of main vibrations: a coefficient of reflection R (intensity), a frequency ν, a width δ in region a frequencies 100–1400 cm^{-1} and compound vibrations in region of high frequencies was studied.

Table 1. Influence of the radiation on a frequency (cm^{-1}) some lines

F, cm^{-2}				
0	$1\cdot10^{19}$	$4\cdot10^{19}$	$5\cdot10^{19}$	$7\cdot10^{19}$
207	206	194	178	—
265	265	264	264	—
357	356	355	354	—
396	396	398	400	—
466	466	462	462	464
697	697	698	690	—

It is shown that neutron irradiation of crystals quartz to integrated fluxes from $1\text{x}10^{19}$ cm^{-2} to $2\text{x}10^{20}$ cm^{-2} strongly influences on their Raman spectra (Fig. 1,2). Radiation effect on the parameters R, ν, δ of row a series of lines in region a frequencies 128...1162 cm^{-1} was determined (Table 1).

For example dose dependence of spectral characteristics for the "soft" modes at 128 cm^{-1} are presented in Fig. 1. It can be seen from Fig. 1 that at the initial stage of irradiation these parameters changes little. A noticeable changes of functions ν(F) and δ(F) occurs for $F>10^{19}$ cm^{-2}, when a rapid phase transition takes place. It is found that the both dose dependencies (Fig. 1) exhibit breaks and then near of some critical point these parameters haven`t registered.

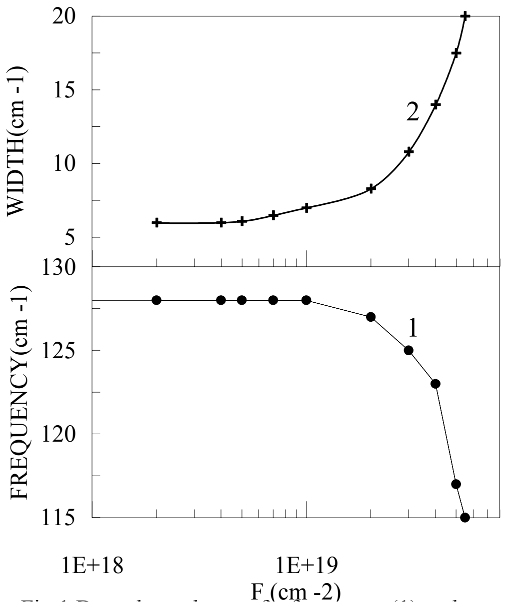

Fig. 1. Dose dependence of a frequency (1) and width (2) for the line 128 (cm^{-1})

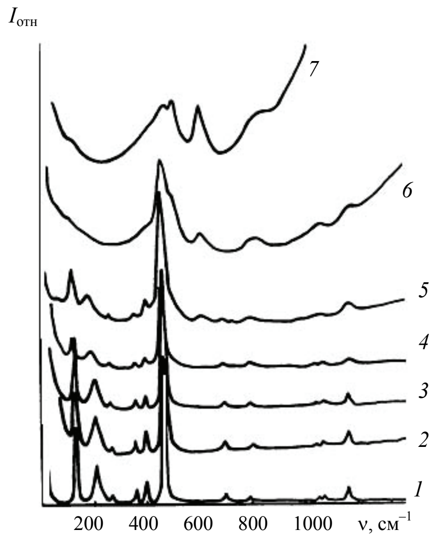

Fig. 2. Raman spectra α-quartz at: $F = 0$ (*1*), $2 \cdot 10^{19}$ (*2*), $3 \cdot 10^{19}$ (*3*), $4 \cdot 10^{19}$ (*4*), $5 \cdot 10^{19}$ (*5*), $7 \cdot 10^{19}$ (*6*) and $2 \cdot 10^{20}$ cm^{-2} (*7*)

Table 2. Radiation changed $R_i(F)$ function for a line 207 cm^{-1}

F [cm^{-2}]	R_j [a.u.]	R_ν [a.u.]	R_δ [a.u.]
0	1,00	-	-
2.x10^{18}	1,00	0,3	5
5x10^{18}	0,98	0,5	9
1x10^{19}	0,98	0,5	22
2x10^{19}	0,82	1,5	36
5x10^{19}	0,50	6,3	81
7x10^{19}	0,00	-	-

A kinetics change of radiation effects in dependence of F for the intensity (R_j), frequency (R_ν), width (R_δ) of another "soft" modes – 207 cm^{-1} in Raman spectra is given in the Table 2. It can be seen from Table 2 that a curves $R_i(F)$ to very nonmonotonically are changed in the dose range, where according to XRD data the phase of α – β transition occurs. This is also confirmed by the variation in the structural parameters of the crystals quartz. For instance, according to X-ray diffraction data (XRD) take off on a DRON-3M diffractometer for irradiated at $F=10^{17}–10^{20}$ cm^{-2} the samples, the Y(F) dependence for a some reflections and the lattice parameters are nonmonotonic, with an peculiarity at the critical point (F_c).

Table 3. The dose dependence of structure parameters of the crystals

F [cm^{-2}]	Y_1 [a.u.]	Y_2 [a.u.]	c [nm]
0	4,5	0,2	0,5404
5x10^{18}	4,4	0,2	0,5404
2x10^{19}	4,0	0,5	0,5409
4x10^{19}	3,4	1,6	0,5420
6x10^{19}	2,7	2,1	0,5444
7x10^{19}	2,4	2,4	0,5450

The special behavior of intensity reflections 3301 and $\bar{3}301$ (Table 3 – Y_1 and Y_2), responsible for phase transition visualization, is of particular interest. It was found that $Y_1/Y_2(F) = 1$ and the quartz lattice parameters c(F) = 0,545 nm near F_c = 7x10^{19} cm^{-2} (Table 3) are well as at the temperature of the phase transition (T_c = 573°C). These relations testify about formation finishing of state like the high-temperatures phase at given fluxes (F_c).

The results on both lines (128, 207 cm^{-1}) and for other lines of scattering are analyzed in comparison with the Raman spectra of the single crystal quartz temperature dependence [8]. It is supposed that in α-quartz samples irradiated to fluxes near F_c the β-quartz like structure are formed.

The influence of radiation on a spectral characteristics of quartz crystalline lattice main modes was determined by the infrared spectroscopy methods. In addition, at the intensive irradiation the induced damage of the structure at the phase transition was discovered in the infrared reflection spectrum the critical transformation in the region of E-type the symmetric and antisymmetrical valent vibrations. Besides, as well as disappearance of bands 1180, 697, 128(E) cm^{-1} was discovered, which is correlated with lattice dynamics by heating crystals to T_c (according of calculations Mathieu [9]) and indicated to end of the phase α-β transition at this critical point.

At the intensive phase transition was discovered critical transformation of infrared and Raman spectra in the region of valent and soft modes. A conception of percolation mechanism of radiation-induced the phase α-β transition in the quartz is proposed.

The effect of radiation on the photorefractive, permitivity and polarizability advanced oxides materials have been adequately studied. In this work of two neutron – radiated modifications of the fused quartz: low-temperature of crystalline quartz(I) and quartz glass (II) (silica) were studied. We investigated plates with the some history, pure and geometry. The samples I-II were irradiated with different neutron fluxes in the channels of a WWR-SM reactor.

We measured the electrical parameter- dielectrical permeability (ε) at 1 kHz of neutron-irradiated quartz (1) and silica (11) plates. The ε(F) dependence was found to be nonmonotonic, with a kink near dose $F=4x10^{19}$ cm^{-2} (for samples 1) and $6x10^{19}$ cm^{-2} (for samples 11). Using these data and dose dependence of the refractive index (n), we calculated the total (α_1) and electronic (α_2) polarizabilities of the samples from the Klausius-Mossoti and Lorentz formulas.

Using the available data on the temperature dependence of the dielectrical permeability and refractive index for the crystal, we evaluated these parameters as a function of temperature. Analysis of the α_1(F) and the α_1(T) curves showed the correlation between change of the polarizabilities of crystals at the thermally and radiation treatments (Table 4). The obtained values were revealed an identical these parameters at they at $F=7x10^{19}$ cm^{-2} and T=700 ^{0}C, where according to the measured diffractograms, an intense phase transition of the crystal.

Table 4. The dose and temperature dependence of polarisabilities in the quartz

F, cm^{-2}	α_1, A^3	T, ^{0}C	α_1, A^3
0	4,86	20	4,85
$1x10^{18}$	4,86	100	4,85
$1x10^{19}$	4,85	200	4,86
$3x10^{19}$	4,86	300	4,89
$5x10^{19}$	4,89	500	4,94
$7x10^{19}$	4,99	700	4,99

We measured the photorefractive ability – the refractive index of the light at the wave length λ=360–650 nm of reactor-irradiated plates. The function n_1(F), n_2(F) of a samples I-II dependence was found to be nonmonotonic, with a kink near the high fluences. For example, it is established that at the initial stage ($F=10^{18}$–10^{19} cm^{-2}) of irradiation of the samples these parameters of the crystals and the glass change little (Fig. 3). A noticeable increase them and generation a point defects (E′-centers) occurs at the second stage (for $F>10^{19}$ cm^{-2}, Fig. 3).

It is of particular interest to study the effect of high doses, which the n(F) curves exhibits similar behavior, with an upturn around of a fluence $F=4x10^{19}$ (the samples II) and $9x10^{19}$ cm^{-2} (the samples I). The dispersion curve n(λ) of the refractive index of the light in the indicated limits is reflected the following irradiation in the reactor of the samples. It is established that at those critical doses they are nonlinear changed.

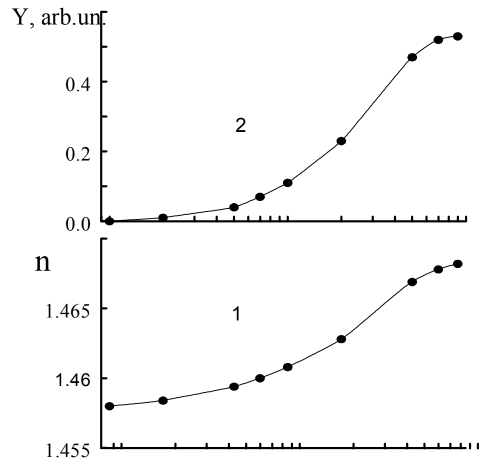

Fig. 3. Dose dependence of radiation defects Y(F)-(2) and n (F)-(1)

Analysis of radiation behaviour of the refraction index for different wave lengths is showed that the irradiation in the first approximation does not lead to any changes in absorption of quartz crystal. Then the observed radiation induced change of photorefraction is likely related to the change of density, which was experimentally observed with the technique of hydrostatic weighing of the irradiated plates. Indeed, the obtained dose dependence ρ(F) is in good correlation with that of n(F) and Y(F) (Fig. 3).

We suppose that the effect change of kink of the n(F) curves peculiarities at the fluences $9x10^{19}$ cm^{-2} and $4x10^{19}$ cm^{-2} is connected with the phase transition in the crystalline quartz and quartz glass accordingly. This is confirmed by X-ray diffraction data. Proceeding from approximation products of radiation changes in photorefractive ability of these oxides materials, a possibility of the percolation mechanism of radiation-induced phase transition in them is discussed.

3. Conclusions

The main results obtained in this work are:

(1) The process of radiation damage and phase transition in alpha-quartz has been determined by spectroscopic methods. It is discovered that dose dependence was changed very nonmonotonicaly for some modes in Raman spectra.

(2) At the intensive phase transition was discovered critical transformation of infrared spectra in the region of valent and compound vibrations and harmonics. A conception of percolation mechanism of radiation-induced the phase transition in the quartz is proposed.

(3) The possibility of radiation modificated some electronic and optical parameters of the solid state dielectric materials by irradiated with a neutrons fluxes have been investigated.

4. References

[1] S. Yoshio, U. Takehio, J. Appl. Phys. 29, 1159 (1990).

[2] K.H. Jong, S. W. McKeever, J. Phys. D. 23, 237 (1990).

[3] S.T. Davey, J.R. Davis, K.J. Roeson, Appl. Phys. Lett. 52, 465 (1988).

[4] K. Fukumi, A. Chagahara, M. Satou et al., J. Appl. Phys. 29, 905 (1990).

[5] K.H. Jong. S.W. Keever, J. Phys. D. 23, 237 (1990).

[6] C.B. Azzoni, F. Meinard, A. Palary, Phys. Rev. B. 49, 918 (1994).

[7] U.B. Potozkiy, M.U. Gorbachuk, Neorgan. Mater. 37, 577 (2001).

[8] P.K. Narayanaswamy, Proc. Ind. Acad. Sci. 28 A, 417 (1948).

[9] J.F. Scott, P. Porto, Phys. Rev. 161, 903 (1967).