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Structural Transformations in Quartz under Neutron Irradiation

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ABSTRACT: The influence of structural inhomogeneities of quartz crystal in the form of β-phases, metamict phases, as well as point defects and impurities on the kinetics of phase transformations occurring in quartz crystals under neutron irradiation has not been sufficiently studied.Therefore, in this paper, the X-ray structural characteristics of quartz crystals grown on seeds irradiated with neutrons at a dose of $5*10^{18}$, 10^{19} and $5*10^{19}$ n/cm^2 after additional neutron irradiation of 10^{16}-$8*10^{20}$ n/cm^2 are investigated. Based on the analysis of X-ray diffraction studies, the nature of intrinsic defects in the structure of quartz crystals, their role in the formation of other structural transformations of quartz crystals has been studied. Based on the analysis of experimental data, it is shown that in crystals grown on neutron-irradiated seedings there is an effect of "radiation heredity"-inheritance of radiation-induced point defects, β-phase and metamict phases of quartz crystals with an overgrown layer.

KEY WORDS: quartz, seed, radiation, neutron, phase transition, phase embryo, radiograph, radiation heredity, diffractometer.

I.INTRODUCTION

This paper presents an analysis of data on the characteristics of Neutron Irradiation. Data analysis for this field shows that the data related to the field has been analyzed in the following literature and data analysis has been carried out [1]. According to the information given in them, it is determined as follows. Crystalline quartz is a polymorphic material. When heated above 573 ° C, quartz enters the β-phase. The temperature α-β transition of crystalline quartz by X-ray diffraction analysis has been studied in detail in [1]. At the same time, it was shown in [2-4] that a structural α-β transition is also observed in crystalline quartz under the action of neutron irradiation. Despite a large number of works devoted to the study of structural changes in crystalline quartz under the influence of neutron irradiation, there is no consensus on the mechanism of phase transition under neutron irradiation to date. In [5], based on the analysis of data from X-ray studies and raman scattering, it is assumed that the structural phase transition in α-quartz under neutron irradiation is due to directional, correlated displacements of atoms and there is a pre-transition state. However, in [2,6], based on theoretical calculations and experimental data, it was shown that the structural phase transition in neutron-irradiated quartz crystals is caused by "displacement peaks". Earlier, in order to clarify the question of the mechanism of phase transition in α-quartz, we determined by X-ray diffraction analysis the amount of β-phase in artificial crystals irradiated with neutrons at a dose of 10^{18}, 5.10^{18} and 10^{19} n/cm^2 and found that the amount of β-phase from the total volume of the crystal is 25, 40, 58%, respectively, that it agrees well with the theoretical calculation.It is shown that the structural α- The β-transition in artificially grown high-purity α-quartz can occur at lower values of the integral neutron flux (10^{18} n/cm^2).At the same time, the volume fraction of the β-phase formed during neutron irradiation increases by accumulation as the dose increases (before the onset of the amorphization stage in irradiated quartz crystals).However, it should be noted that the volume fraction of the high-temperature quartz phase does not increase proportionally to the integral neutron flux, but has a more complex character

II.THE METHOD OF THE EXPERIMENT

In this work, X-ray diffraction studies of artificial quartz crystals irradiated with neutrons up to a certain dose and crystals grown on seedings previously irradiated with an integral neutron flux of 5.10^{18}, 10^{19} and 5.10^{19} n/cm^2 were carried out.

X-ray images of the samples were taken on a DRON-2.0 diffractometer using Cu-Ka radiation (λ =0.1542 nm). The powder sample was poured into a standard 0.5 mm deep cuvette.

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III.THE RESULTS OF THE EXPERIMENT

Table 1.1. shows the measurement results for two reflections of quartz crystals irradiated with neutrons and grown on neutron-irradiated seedings . After the first measurement, the samples irradiated with neutrons were measured a second time (II) after 120 days with full compliance with the experimental conditions. The table shows that the intensity and angular position of diffraction reflections varies. Of these, the structural reflex (101) belongs simultaneously to the α- and β-phase of quartz. The calculated peak intensity (101) for the α- and β-phases is almost the same, so the intensities of the remaining diffraction reflections can be tied to it. As the radiation dose increases, the angular position of the peak (101) shifts towards a smaller scattering angle. A similar displacement of the angular position of the reflection (101) in the same direction was observed in experiments investigating the α-β transition of quartz under heating [1]. But the amount of displacement during irradiation was 2/3 of the temperature displacement. The intensity of diffraction reflection (111), belonging only to the a-phase of quartz, decreases with an increase in the integral neutron flux, with the exception of the sample irradiated with a dose of 10^{19} n/cm^2 I measurement.For (111), a displacement of the location in the direction is also observed during neutron irradiation, as for (101).

IV.DISCUSSION OF THE RESULTS

These results show that in quartz crystals, the observed α-β transition under neutron irradiation has a similar structural character as the transition under heating. However, the transition under neutron irradiation is not completed completely. This is evidenced by the insufficient displacement of diffraction reflections towards a smaller scattering angle, a small change in the parameters of the unit cell and X-ray density (Table 1.2) and incomplete disappearance of the structural reflex (111). It is interesting to note that during repeated measurement of (II) samples irradiated with neutrons at a dose of 10^{17} and 5.10^{18} n/cm^2, a threefold increase in the intensity of diffraction reflection (111) is observed, i.e. an increase in the α-phase after the I-th measurement. For a sample of 5. 10^{19} n/cm^2, the increase in intensity is not fixed. Apparently, this is due to the strong vibrational excitation of the structural units of quartz crystals- tetrahedra in the dose range 10^{17}-5. 10^{18} n/cm^2. At these doses, silicon atoms, shifting towards the structural channel of quartz, somewhat reorient the silicon-oxygen tetrahedra. But this reorientation of tetrahedra is reversible, since the energy resulting from the distortion of the lattice is still overcome by silicon atoms in the state of excitation. At an irradiation dose of 5.10^{19} n/cm^2, the lattice distortion energy will increase so much that repeated reorientation of tetrahedra becomes impossible. It is interesting to note that when (II) samples irradiated with neutrons at a dose of 10^{17} and 5.10^{18} n/cm^2 are re-measured, a threefold increase in the intensity of diffraction reflection (111) is observed, i.e. an increase in the α-phase after the I-th measurement. For a sample of 5. 10^{19} n/cm^2, the increase in intensity is not fixed. Apparently, this is due to the strong vibrational excitation of the structural units of quartz crystals- tetrahedra in the dose range 10^{17}-5. 10^{18} n/cm^2. At these doses, silicon atoms, shifting towards the structural channel of quartz, somewhat reorient the silicon-oxygen tetrahedra. But this reorientation of tetrahedra is reversible, since the energy resulting from the distortion of the lattice is still overcome by silicon atoms in the state of excitation. At an irradiation dose of 5.10^{19} n/cm^2, the lattice distortion energy will increase so much that repeated reorientation of tetrahedra becomes impossible. It is known that the uneven occurrence of impurities in individual pyramids and growth zones, as well as radiation-induced defects [7] lead to a change in the parameters of the quartz crystal lattice.
In [4,6-8], quartz crystals grown on neutron-irradiated seedings with a dose of 10^{18}, 5.10^{18}, 10^{19} and 5.10^{19} n/cm^2 were studied. It was shown that in a quartz crystal grown on a seed irradiated with a neutron flux of less than 10^{18} n/cm^2, no significant changes were observed in the overgrown layer. However, cracks are observed in crystals grown on seeds irradiated with a dose of more than 10^{18} n/cm^2, and with an increase in the neutron dose, its increase is detected. The appearance of cracks in [8-9] is caused by the discrepancy between the parameters of the crystal lattice of the seed and the crystal layer that has grown on it. As a result of this discrepancy between the seed and the overgrown layer, a stress is created that causes fracturing of the grown crystal. As shown in [8-10,11], a neutron-irradiated crystal consists of a mixture of high-temperature and low-temperature modifications of quartz. Therefore, it was of interest to study quartz crystals grown on a neutron-irradiated seed. At the same time, it was assumed that if there are beta-phase nuclei in a neutron-irradiated seed, then the beta-phases of quartz should also be observed on the crystal layer that has grown on it.

To verify this assumption, the structural changes of artificial quartz crystals grown on neutron-irradiated seedings were studied. The experimental results are given in Table 1.1 and compared with the data of a neutron-irradiated

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quartz crystal. Table 1.1 shows that in crystals grown on neutron-irradiated seeds, with increasing dose, the same pattern is observed as in crystals irradiated with neutrons.

V. CONCLUSION

Based on experimental data, it is shown that all the structural characteristics of the seed are transferred to the grown crystal. This opens up the possibility of obtaining quartz crystals with predetermined properties. Diffraction parameters of quartz irradiated by neutrons and grown on neutron-irradiated seedings (n/cm^2).

Table 1.1

Sample	measurement	HKL	10^{17}		5.10^{18}		10^{19}		5.10^{19}	
			J	2θ	J	2θ	J	2θ	J	2θ
Neutron-irradiated crystals	I	101 111	1,00 4,00	$26^0$45’ $40^0$22’	100 3,12	$26^0$42’ $40^0$20’	100 6,41	$26^0$38’ $40^0$15’	100 2,89	$26^0$33’ $40^0$12’
	II	101 111	100 11,49	$26^0$45’ $40^0$22’	100 10,33	$26^0$40’ $40^0$18’	100 2,72	$26^0$36’ $40^0$12’	100 3,16	$26^0$34’ $40^0$11’
Crystal grown on neutron-irradiated seed	III	101 111	- -	- -	100 4,51	$26^0$39’ $40^0$17’	100 3,06	$26^0$36’ $40^0$12’	100 2,36	$26^0$34’ $40^0$12’

Unit cell parameters and density of the studied quartz crystals

Table 1.2

Measurement	Sample	a (nm)	c (nm)	c/a	$\rho_p(10^3 kg/m^3)$
I	10^{17}	$0,491_2$	$0,540_2$	$1,099_6$	$2,67_3$
	5.10^{18}	$0,491_5$	$0,539_0$	$1,099_6$	$2,67_6$
	10^{19}	$0,492_1$	$0,541_1$	$1,099_6$	$2,66_1$
	5.10^{19}	$0,492_6$	$0,541_1$	$1,099_6$	$2,65_9$
II	10^{17}	$0,491_4$	$0,540_9$	$1,100_7$	$2,66_7$
	5.10^{18}	$0,491_7$	$0,539_5$	$1,097_2$	$2,67_1$
	10^{19}	$0,492_0$	$0,539_9$	$1,097_3$	$2,66_6$
	5.10^{19}	$0,492_9$	$0,541_6$	$1,098_9$	$2,64_5$
III	5.10^{18}	$0,491_4$	$0,540_0$	$1,098_9$	$2,67_1$
	10^{19}	$0,491_4$	$0,540_2$	$1,099_1$	$2,67_1$
	5.10^{19}	$0,491_5$	$0,540_2$	$1,097_2$	$2,67_0$

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