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Journal of Nuclear Materials 273 (1999) 228–231

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Letter to the Editors

Irradiation swelling of explosively shocked materials

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Received 27 November 1996; accepted 10 February 1999

Abstract

The measurement of AlN, ZnO, $GdAlO_3$ and $Gd_2Ti_2O_7$ swelling after irradiation to 1.1×10^2 n/cm^2 at a temperature <120°C was conducted by X-ray diffraction techniques. Powder specimens were irradiated in the initial state and after explosive shock wave in the pressure range 12–70 MPa. A 25–70% decrease in 'X-ray swelling' was observed in all cases.

1. Introduction

The limited choice of non-metallic materials with thermal, nuclear and other properties making them unique candidates for structural application under irradiation has led us to investigate ways of diminishing their radiation damage by appropriate microstructural changes. The actual task consisted in estimating the effect of explosive shock waves on the decrease in the 'X-ray swelling' magnitude of non-metallic materials belonging to two classes. Zinc oxide and aluminum nitride are materials which have temperature phase transformations of the reconstructive type. They have a structural analogy with beryllium oxide – wurtzite type. AlN is used as a base of the magnetic gauge of ITER diagnostic system. Aluminum gadolinium oxide $GdAlO_3$ (perovskite type) and gadolinium titanium oxide $Gd_2Ti_2O_7$ (pyrochlore type cubic structure) are materials which have temperature transformation of the displacement type, and they are used as neutron absorbing materials.

Rapid displacements of matter accompany explosive shock waves going through the crystal. The structural disorder, grinding of grains up to 100 Å in size [1] in addition to defect production (dislocation loops, vacancies, interstitials) are results of this. The number of defects created depends on the parameters and process of explosive shock waves and on the materials' physical properties. The use of non-metallic materials possessing high defect concentration in their atomic structure can change substantially the kinetics of radiation damage transformations [2]. Consequently, we can expect a decrease in irradiation swelling and the results of this study on materials explosively shocked are presented hereafter.

2. Explosive shock wave treatment of the investigated materials

Initial materials were subjected to an explosive shock wave in the pressure range 12–70 MPa. This treatment has been made in cylindrical ampoules storage. The shock wave was generated in the specimens by explosive charges of coaxial symmetry, which wear in intimate contact either with ampoule wall or using a metal striker in the form of a cylinder. The shock wave conditions were selected with regard to the maximum possible number of defects produced by the explosion and the technological features of the experiment.

3. Irradiation conditions and techniques of investigation

Powder materials were irradiated in the leak-tight ampoules having an inner diameter of 6.9 mm. They

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00049-5

were located in the water of the primary circuit of the SM-reactor. The coolant temperature was 90°C and the fast and thermal neutron fluences reached 1.1×10^{21} n/cm^2 ($E > 0.1$ MeV) and 1.3×10^{22} ($E < 0.68$ eV), respectively. The initial and the explosively shocked samples were irradiated simultaneously.

X-ray investigations have been performed on the diffractometer DRON-2.0 for non-irradiated powders and on the remotely operated diffractometer DARD for the irradiated materials. Cu-K_α radiation was used in both cases. All angular measurements were calibrated with the use of the standard-diamond powder.

4. Experimental results

4.1. Zinc oxide

The explosive shock wave made negligible changes in the width of diffraction lines, which were only visible at the large angles. Lattice parameters were measured from the position of all the {h k 0} and {0 0 l} diffraction peaks for non-irradiated materials. They are $a_0 = 3.2492 \pm 0.0002$ Å and $c_0 = 5.2060 \pm 0.0007$ Å for starting samples and $a_{exp} = 3.2499 \pm 0.0003$ Å; $c_{exp} = 5.2033 \pm 0.0008$ Å for the explosively shocked ones. The corresponding cell volumes are $V_0 = 47.60 \pm 0.01$ Å^3 and $V_{exp} = 47.58 \pm 0.01$ Å^3.

Since irradiation of the wurtzite crystals (ZnO as well as AlN and BeO) is the cause of diffuse scattering at (h k l)-lines ($l \neq 0$) [3,4], moved at greater angles than Bragg's angles, the parameter a was measured at (h k 0) lines, and c was measured at (h k l) lines, ($l = 1$). At the same time the Bragg's component could be separated at (0 0 2) line in the material after explosive treatment (Fig. 1). The results of measurements are given in Table 1 .

The volume of lattice $V_{0.irr}$ is equal to (47.73 ± 0.12) Å^3; $V_{exp.irr}$ is equal to (47.90 ± 0.12) Å^3. Therefore the value of X-ray swelling for initial materials is equal to 0.6% and 0.3% for explosive wave shocked materials.

4.2. Aluminum nitride

The diffraction lines widen more than for ZnO ones (by 30–40% for the first lines and by 100% at higher angles) after explosive treatment (see Fig. 2). The accuracy of measurement was negligible at angles more than 95°C. The lattice parameters determined from all lines were $a_0 = (3.1152 \pm 0.0002)$ Å, $c_0 = (4.9930 \pm 0.0008)$ Å, $a_{exp} = (3.1194 \pm 0.0004)$ Å, $c_{exp} = (4.987 \pm 0.001)$ Å. The elementary cell volumes corresponding to the initial and explosively shocked materials are $V_0 = (41.96 \pm 0.01)$ Å^3 and $V_{exp} = (42.02 \pm 0.02)$ Å^3, respectively.

As for ZnO crystals, (h k l) lines with $l > 1$ could not be used for determining lattice parameters of irradiated AlN. Moreover, the lattice parameter c could not be deduced from the (0 0 2) peak position. The results of lattice parameter measurements of irradiated AlN are given in Table 2. The volume of ZnO lattice $V_{0.irr}$ is

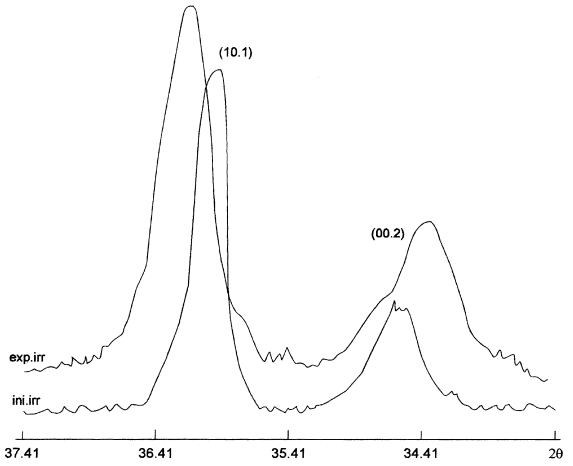

Fig. 1. Detail of diffractograms of irradiated ZnO.

Table 1
The measured lattice parameters of irradiated samples of ZnO

h k l	Initial		After expl.	
	2θ (deg.)	a and c (Å)	2θ (deg.)	a and c (Å)
1 0 0	31.78	3.251 ± 0.004	31.77	3.252 ± 0.004
0 0 2	34.43		34.41 [a]	5.212 ± 0.006
1 0 1	36.23	5.231 ± 0.006	36.25	3.253 ± 0.004
1 0 2	47.64		47.52	
1 1 0	56.62	3.251 ± 0.002	56.67	3.248 ± 0.002
1 0 3	62.82		62.89	
2 0 0	66.38	3.252 ± 0.002	66.38	3.252 ± 0.002
1 1 2	67.95		67.97	
2 0 1	69.09	5.230 ± 0.003	69.12	5.212 ± 0.003

[a] Bragg's maximum.

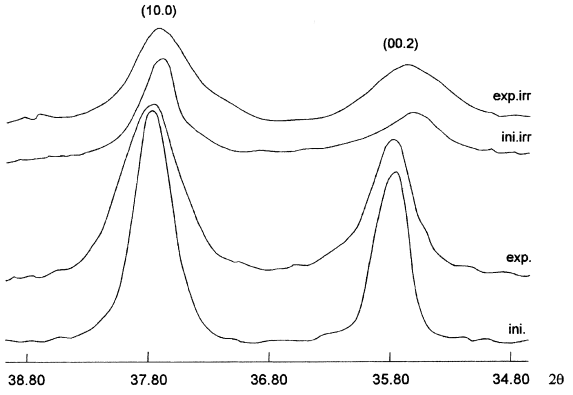

Fig. 2. Detail of diffractograms of AlN materials.

Table 2
The measured lattice parameters of irradiated samples of AlN

h k l	Initial		After expl.	
	2θ (deg.)	*a* and *c* (Å)	2θ (deg.)	*a* and *c* (Å)
1 0 0	33.16	3.120 ± 0.004	33.15	3.121 ± 0.004
0 0 2	35.86		35.88	
1 0 1	37.78	5.054 ± 0.006	37.80	5.021 ± 0.006
1 0 2	49.66		49.68	
1 1 0	59.24	3.118 ± 0.002	59.23	3.120 ± 0.002
1 0 3	65.88		65.90	
2 0 0	69.62	3.119 ± 0.002	69.91	3.119 ± 0.002
1 1 2	71.22		71.23	
2 0 1	72.44	5.053 ± 0.003	72.46	5.025 ± 0.003

equal to (42.57 ± 0.18) Å^3; $V_{exp.irr}$ is equal to (42.22 ± 0.19) Å^3. So the value of X-ray swelling for initial material is equal to 1.4% and 0.5% for the explosively shocked material.

4.3. Aluminum gadolinium oxide

Wurtzite crystals of ZnO and AlN did not undergo structural changes after explosive wave treatment and irradiation. Since aluminum gadolinium oxide presents a phase transformation of shift type, these treatments induce phase transformations. This results in the weakening and disappearance of some lines. The characteristics of the diffraction lines are given in Table 3.

It is known that the perfect structure of $GdAlO_3$ is difficult to obtain. The analysis of X-ray diffraction patterns of initial materials for D_{2h}^{16} structure gives the following values relative to CARD ASTM 9–85: $a = 5.256 \pm 0.002$ Å, $b = 5.304 \pm 0.002$ Å, $c = 7.437 \pm 0.003$ Å.

The explosive wave treatment changed the structure of the material. The irradiation modified it further. In order to estimate the X-ray swelling, we compared interplanar spacing deduced from only lines present on all four X-ray patterns: $d_0 = 2.637$ Å; $d_{exp} = 2.630$ Å; $d_{0.irr} = 2.670$ Å; $d_{exp.irr} = 2.654$ Å.

Deciding the $GdAlO_3$ crystals is isotropic, the explosive wave treatment decreases the crystal volume by ~0.2%, X-ray swelling of initial material is equal to +3.7%, and it is +2.7% for explosive wave shocked material.

4.4. Gadolinium titanium oxide

The explosive wave treatment distorted the crystalline lattice and partially disordered the structure. All diffraction peaks became wider; the width of the (6 6 2) line increased twofold. The intensity of lines of all samples decreased significantly after irradiation. The characteristics of some diffraction lines of $Gd_2Ti_2O_7$ are given in Table 4. The diffraction peaks of irradiated materials were not found for $2\theta > 60°$. The lattice parameter determined from all the lines was $a_{exp} = 10.195 \pm 0.002$ Å. The level of discord of the structure may be estimated from the ratio of the total intensity to the intensity of substructure lines. Initially it was 5.0 and after explosive wave treatment it was 6.5 (for the ordered structure of pyrochlore it is 4.0).

After irradiation, both the initial material and the explosive wave shocked material are assumed to have a fluorite type structure. Their lattice parameters are: $a_{ini.irr} = 5.099 \pm 0.003$ Å, $a_{exp.irr} = 5.117 \pm 0.003$ Å. Hence the X-ray swelling is equal to 1.3% for initial

Table 3
The characteristics of the diffraction peaks of $GdAlO_3$ samples

h k l Theor.	Initial		Exp.		Initial irrad.		Exp. irrad.	
	2θ (deg.)	*I* imp/s	2θ (deg.)	*I* imp/s	2θ (deg.)	*I* imp/s	2θ (deg.)	*I* imp/s
1 1 0, 0 0 2	23.84	320	23.90	105	–	–	23.68	14
1 1 1	26.71	55	–	–	–	–	–	–
0 2 0	–	–	–	–	–	–	–	–
1 1 2, 2 0 0	34.00	760	34.09	220	33.56	24	33.65	29
0 2 1	36.07	17	–	–	36.20	11	–	–
1 0 3, 2 1 1	40.12	8	40.31	8	–	–	–	–
0 2 2	41.84	78	41.90	63	–	–	–	–
2 0 2	42.07	75						
1 1 3	–	–	–	–	–	–	–	–
2 2 0, 0 0 4	48.82	320	48.83	87	–	–	–	–
0 2 3, 2 2 1	50.38	30	50.41	26	–	–	–	–
2 1 3, 3 0 1	53.61	12	–	–	–	–	–	–
1 1 4, 3 1 0	55.09	62	55.18	48	–	–	–	–
1 3 1	56.27	38	56.38	21	–	–	–	–

Table 4
The characteristics of the diffraction peaks of $Gd_2Ti_2O_7$ samples

h k l	Initial		Expl.		*h k l*	Initial irrad.		Expl. irrad.	
	2θ (deg.)	*I* imp/s	2θ (deg.)	*I* imp/s		2θ (deg.)	*I* imp/s	2θ (deg.)	*I* imp/s
1 1 1	15.12	62	15.06	19	–	–	–	–	–
3 1 1	29.16	73	29.06	40	–	–	–	–	–
2 2 2	30.50	780	30.38	313	1 1 1	30.36	23	30.25	20
4 0 0	35.36	196	35.22	97	2 0 0	35.20	10	–	–
3 3 1	38.64	206	38.48	63	–	–	–	–	–
5 1 1	46.48	83	46.28	25	–	–	–	–	–
4 4 0	50.86	493	50.66	190	2 2 0	50.66	16	50.44	12
5 3 1	53.38	85	53.14	22	–	–	–	–	–
6 2 2	60.48	420	60.20	153	3 1 1	60.19	11	59.96	12
4 4 4	63.48	104	63.18	32	–	–	–	–	–
5 5 1	65.66	32	65.36	11	–	–	–	–	–
7 3 1	71.34	34	71.02	11	–	–	–	–	–
8 0 0	74.80	50	74.44	27	–	–	–	–	–
6 6 2	82.88	191	82.47	58	–	–	–	–	–

materials and 1.1% for explosive wave shocked materials.

5. Conclusion

Results obtained in this work support the hypothesis that the irradiation-induced volume increase of non-metallic materials can be reduced by the use of an explosive shock wave. Evidently, X-ray investigations which have been performed on the diffractometer are not sufficient to analyze completely the imperfect structure of these materials. Nevertheless, using the diffraction line widening we can draw some conclusions about the existence of dislocation structures and grain fragmentation. Structural defects which have appeared as a result of the shock wave are sinks for the elimination of primary radiation defects (Frankel's pairs) and thus reduce the number of stable defects accumulating in the material which results in a swelling decrease.

Explosive shock wave will probably become a universal way to produce swelling resistant non-metallic materials.

Acknowledgements

The work was executed under the support of the Russian Fund of Fundamental Investigation. Grant 96-02-16665.

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