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Physica B 234–236 (1997) 405–406

PHYSICA B

Structure of amorphous Al_2O_3

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Abstract

The structure of amorphous Al_2O_3 samples, prepared by anodic oxidation of aluminium foils, was investigated by X-ray and neutron diffraction. The partial pair correlation functions were determined from computer simulation of the structure using the Reverse Monte Carlo method with the result that amorphous Al_2O_3 is built up by AlO_4 tetrahedra with corner sharing oxygen atoms. Pores in the material were characterized by neutron small-angle scattering.

Keywords: Amorphous materials; Diffraction; Computer modelling

1. Experimental

Amorphous Al_2O_3 samples were produced by anodic oxidation of Al foils as described in a previous paper [1]. In the as-prepared state they contained about 15 at% H_2O and about 3 at% sulfur. From X-ray and neutron diffraction (with the D4B instrument at ILL, Grenoble) the total pair correlation functions were derived, using the methods as described e.g. in Ref. [2]. Small-angle scattering measurements were done with the D17 instrument at ILL.

2. Results and discussion

Fig. 1 shows the neutron diffraction cross sections for an as-prepared sample and after annealing at 300°C and 500°C. The as-prepared sample exhibits a strong inelastic Q-dependent scattering contribution due to the hydrogen atoms. At 300°C the water absorbed in the pores is released and above 450°C also the water of constitution. For the 500°C-sample the decline of $d\sigma/d\Omega(Q)$ with increasing Q follows the theoretical Placzek correction term of Al_2O_3 without a hydrogen contribution.

The experimental total pair correlation functions $G_x(R)$ and $G_n(R)$ are plotted in Fig. 2(a) together with the model curves as obtained by using the reverse Monte Carlo (RMC) computer simulation method [3]. In the RMC calculation an amount of 3.4% sulfur atoms was included. We note that the small peak around $R = 1.5$ Å belongs to the partial S–O correlation function. The three RMC partial pair correlation functions G_{AlAl}, G_{OO} and G_{AlO} are shown in Fig. 2(b) (where $G_{ij}(R) = 4\pi R[\rho_{ij}(R)/c_j - \rho_0]$, c_j is the atomic fraction of component j, $\rho_{ij}(R)$ the number of j atoms per unit volume around i atom, and ρ_0 the mean number density). The average atomic distances, the widths of the peaks and the coordination numbers are listed in Fig. 2(b). The very sharp Al–O peak together with the coordination number $Z_{AlO} = 4.1$ shows that

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PII S0921-4526(96)01044-7

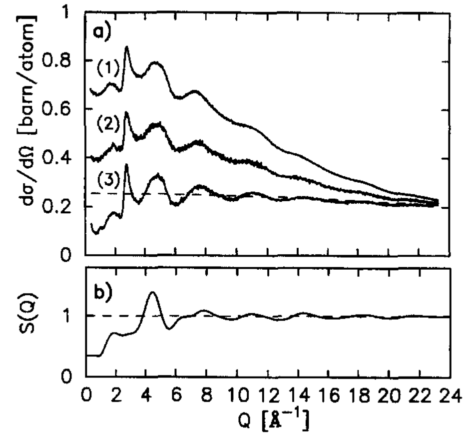

Fig. 1. Amorphous Al_2O_3: (a) neutron diffraction, cross section. (1) as-prepared, (2) 300°C/3 h, (3) 500°C/3 h, --- Placzek correction term for (3). (b) X-ray diffraction, structure factor $S(Q)$.

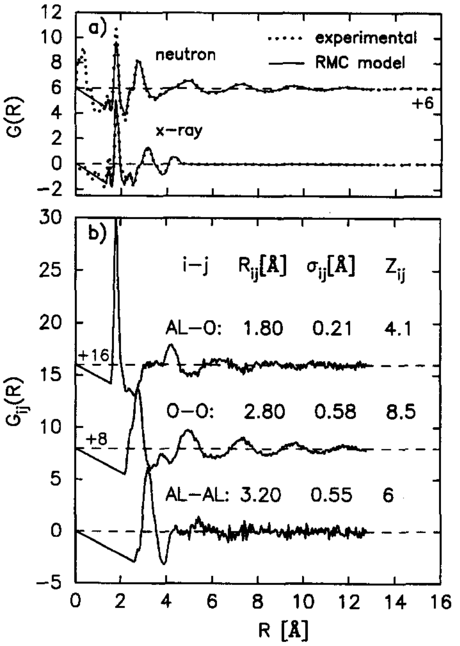

Fig. 2. Amorphous Al_2O_3: (a) total pair correlation functions. (b) partial pair correlation functions from RMC model; atomic distances R_{ij}, widths of pair distributions σ_{ij} and coordination numbers Z_{ij} (j around i).

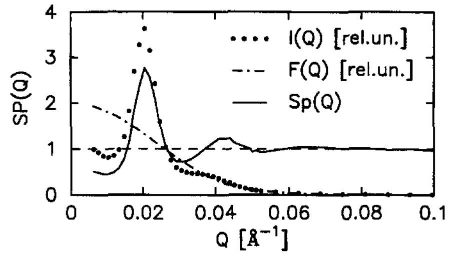

Fig. 3. Amorphous Al_2O_3: small-angle neutron scattering curves, see text.

the amorphous structure is built up by AlO_4 tetrahedra, where $R_{AlO} = 1.80$ Å, and that the number of AlO_6 octahedra, if present at all, must be very small. The distribution of the Al–O coordination number in the RMC cluster can be characterized by the frequencies: $Z_{AlO} = 3(20\%)$, $4(56\%)$, $5(22\%)$. The experimental O–O distance at $R_{OO} = 2.80$ Å is somewhat smaller than the distance 2.94 Å between the O atoms calculated for a tetrahedron where $R_{AlO} = 1.80$ Å. The average O–O distance in the RMC cluster between O-atoms which belong to the same tetrahedron is $R_{OO} = 2.91$ Å. From R_{AlO} and R_{AlAl} the Al–O–Al bond angle between two corner sharing tetrahedra results as 125°. The extended oscillations in the $G_n(R)$ curve in Fig. 2(a), up to at least 16 Å, are mainly due to the well-defined O–O correlations, as can be seen from the partial $G_{ij}(R)$ in Fig. 2(b).

The small-angle neutron scattering effect of as-prepared Al_2O_3, $I(Q)$ in Fig. 3, is caused by pores in the samples. By fitting a form factor $F(Q)$ to this curve the structure factor of the distance correlations between the pores, $\mathrm{Sp}(Q) = I(Q)/F(Q)$, was obtained. The analysis of the data yielded a volume fraction of 8% pores with diameters of about 140 Å at an average distance of 340 Å.

References

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