Local Structure of Radiation Damaged $ZrSiO_4$

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Natural zircon, $ZrSiO_4$, is known to undergo amorphization as a consequence of the α-decay of radioactive impurities. Understanding the structure of radiation-damaged (metamict) zircon is important, since materials with the structure of zircon are used for the immobilization of excess weapons plutonium and other radionuclides. The structure of slightly damaged zircon has already been determined.[1] The structure of heavily damaged $ZrSiO_4$ is, however, not known. We approached the problem by using high-energy x-ray diffraction and the atomic pair distribution (PDF) technique. The experiments were carried out at the SRI-CAT 1-ID beamline at the Advanced Photon Source, Argonne, using x-rays of energy 80.6 keV. Three samples were studied: perfectly crystalline zircon, sol-gel derived amorphous zircon and heavily damaged natural (metamict) zircon. The experimental structure factors, S(Q), are shown in Fig. 1 and the corresponding atomic pair distribution functions (PDF), G(r), in Fig. 2.

As can be seen, the structure factor for metamict $ZrSiO_4$ has only a few broad features, a characteristic of a completely amorphous material. Interestingly, the structure factor of amorphous $ZrSiO_4$ obtained by the sol-gel technique and that of metamict zircon are not quite the same, showing that the two materials do not have the same local atomic ordering. Currently, the details of the atomic ordering in metamict zircon are being explored by reverse Monte Carlo modeling of the diffraction data. The results will be reported elsewhere.

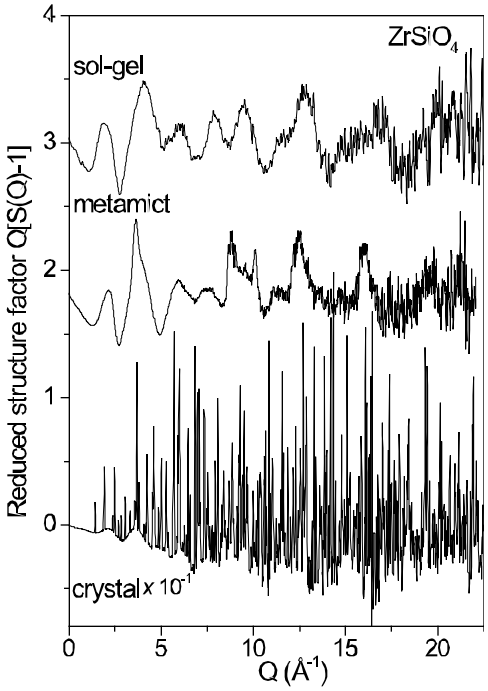

FIG. 1. Experimental structure factors for crystalline, sol-gel derived and metamict zircon.

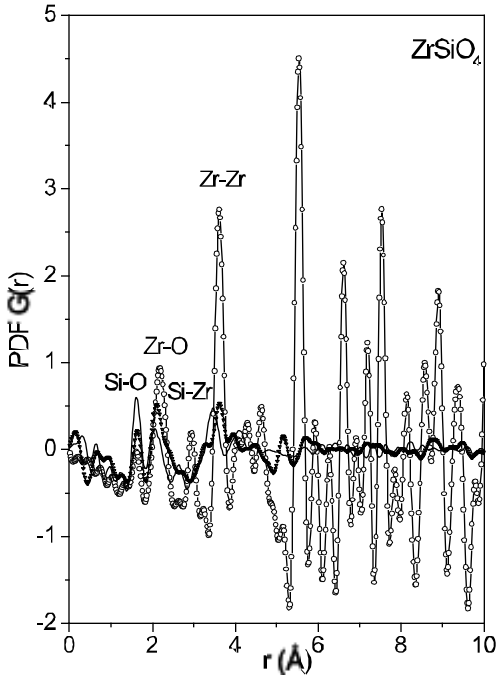

FIG. 2. Experimental PDFs for crystalline (open symbols), sol-gel derived (thin line) and metamict zircon (thick line). Peaks are labeled with the corresponding atomic pairs.

References

[1] S. Rios, Th. Malcherec, E. Salje and Ch. Domeneghetti, Acta Cryst. B **56**, 947 (2000).

Acknowledgments

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