The Evaluation of the Thermal Conductivity of Hyperstoichiometric UO_{2+x} by Molecular Dynamics Simulations

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Uranium dioxide, UO_2, has been the most widely used nuclear fuel in light water reactors. In the fuels, released oxygen molecules oxidize the UO_2 fuel itself by fission, and, consequently, raise the ratio of oxygen to uranium (O/M = 2x). The O/M ratio significantly affects thermal properties, e.g. melting point and thermal conductivity, and this effect has not been studied sufficiently for $x > 0$ experimentally or with simulations.

In the present study, the thermal conductivity of hyperstoichiometric UO_{2+x} has been investigated by the equilibrium molecular dynamics (EMD) simulation at temperatures up to 2000 K. The interatomic potential used was the Born-Mayer-Huggins (BMH) type with the partially ionic model [1]. The thermal conductivity was determined using the transport coefficients of the phenomenological equations [2]. According to the Green-Kubo relationship, such transport coefficients were defined as the integration of the correlation function of the energy and charge fluxes.

In the U-O system, hyperstoichiometric UO_{2+x} exists homogeneously at temperatures above 600 K, and has the fluorite structure. The BMH potential parameters for UO_{2+x} were determined using thermal expansion data [3]. The results obtained by the EMD calculations were: (1) cross coupling effects between energy and charge fluxes were observed at large x values and high temperatures; (2) with the increase of x, the thermal conductivity decreased and its temperature dependence was reduced, which was attributed to phonon scattering by excess oxygen.

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