Energetics and Annealing Behavior of Radiation Damage in Pyrochlores

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Pyrochlores, ordered structures derived from the fluorite aristotype, have been proposed as waste forms for actinides and fission products. Their susceptibility to radiation damage varies strongly with composition and disorder can also be induced by grinding, low temperature synthesis, and in some cases, heating. Swift heavy ion irradiation can now produce macroscopic amounts of sample with controlled and uniform degrees of damage. Thermodynamic study of radiation damage damage and its annealing by high temperature calorimetry combined with neutron scattering and X-ray diffraction of such materials gives critical insights of their energetics and annealing behavior. We report a pilot study of swift heavy ion irradiation $Dy_2Ti_2O_7$ phyrochlore using a combination of techniques. The energetic difference between the irradiation amorphized and unirradiated ordered samples was probed by high temperature oxide melt solution calorimetry. Substantial energetic destabilization of the irradiated $Dy_2Ti_2O_7$ sample relative to ordered pyrochlore by 243 kJ/mol is revealed. The amount of heat release and the temperature region of annealing events were measured by differential scanning calorimetry. The total annealing enthalpy is -137.3 kJ/mol, which is about half of the energetic difference between the ordered and amorphized pyrochlore. Annealing back to a fully ordered structure energetically equivalent to the starting material appears to be a complex multistep process, perhaps involving different kinetics for ordering on the cation and anion sublattices.