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Method of SHS Processing the Reactor Graphite Waste Including Fuel Fragments: Thermodynamic and Experimental Modeling

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During an operation period of uranium-graphite reactors, reactor graphite waste accumulates in bulk-form (fines and crumbles of graphite, fuel, and fuel assemblage fragments). Under neutron irradiation the long-lived radionuclides of actinides U, Np, Pu, Am, Cm and others elements (¹⁴C, ⁶⁰Co, ⁶³Ni, ⁹⁰Sr, ¹³⁷Cs, ¹⁵¹Sm, ¹⁵⁴Eu and the like) are expected to be formed in the waste. When decommissioning uranium-graphite reactors, the reactor graphite waste is to be reliably isolated from the environment. The proposed reactor graphite waste processing is based on a self-sustaining high-temperature synthesis (SHS) reaction yielding stable ceramics including radionuclides, particularly biologically hazardous carbon ¹⁴C, in the chemically bound state. Thermodynamic simulation and experimental investigation of the SHS processing of the reactor graphite waste are performed within a wide range of starting mixture compositions. Lanthanum, cerium, neodymium, samarium, and gadolinium oxides have been used as substituents of the actinides. In the course of SHS processing, the reactions proceed to result in the formation of a titanium carbide - corundum ceramic matrix and lanthanide aluminate perovskites. X-ray diffraction analysis of the composition of the SHS reaction condensed products and chemical analysis of the gas phase composition have been carried out. The results obtained correlate well with the thermodynamic simulation data. The efficiency of an SHS process for immobilization of the irradiated reactor graphite waste containing lanthanide oxides has been demonstrated.