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On immobilization of high-level waste in an Y–Al garnet-based cermet matrix in SHS conditions

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Abstract

A method of high-level waste (HLW) radionuclide immobilization in a long-life matrix based on Y–Al garnet, a material highly chemically resistant to natural environments, has been developed for the ultimate HLW isolation from the environment. Model systems containing Ce, Nd, Sm, Zr, Mo, ^{238}U, and ^{241}Am were used in the study as simulators of HLW radionuclides. An energy-saving technology of self-propagating high-temperature synthesis (SHS) was employed to synthesize the matrix material with fixation of HLW radionuclide simulator elements in the Y–Al garnet structure. The results of an X-ray phase analysis for the synthesized materials have shown that the simulator elements, as well as uranium and americium are incorporated strongly in the structure of Y–Al garnet predominantly forming the matrix's major neoformation fit for environmentally safe disposal. The produced synthetic mineral-like matrices feature high water resistance, the property confirmed by a very low rate (10^{-9}–10^{-10} g/cm^2·day) of americium leaching into water. Besides high strength of the americium fixation in the structure of Y–Al garnet, the latter's carryover is small at high temperatures due to a short duration of the process.

The technically, economically and environmentally attractive novel HLW immobilization technique proposed herein may form the basis for the closing process of the spent nuclear fuel reprocessing.

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The new technology platform of nuclear power based on a closed nuclear fuel cycle (CNFC) suggests the need for new technologies to be created to reprocess spent nuclear fuel (SNF). The progressing accumulation of SNF reprocessing products (high-level waste) poses a major danger to human activities and the environment and hinders the evolution of nuclear power. The HLW formation requires the problem of the HLW conditioning with the absolute waste isolation from the biosphere to be solved. One of such possible ways is to immobilize HLW in matrix materials – highly durable analogs of natural minerals chemically resistant to natural water environments. The key to the effective application of such matrix materials is the relative simplicity of the chemical processes involved in the synthesis thereof, which may be decisive when it comes to the selection of the proper commercial technology.

No final decision has been so far made as part of the CNFC as to the preferred SNF reprocessing technology and concerning the selection of the matrix material for the HLW isolation.

The most common of the stable matrix materials are mineral-like oxide ceramic neoformations obtained in solid-phase high-temperature chemical processes. These processes form the closing HLW handling phase.

As far as water-based treatment processes are concerned, the most promising of these appears to be the one with radionuclides of selected groups extracted from liquid HLW, fixed on sorbents and incorporated at a high temperature into a crystalline structure of synthesized mineral-like formations. The techniques used at the final HLW conditioning stage may include cold compaction followed by high-temperature baking (CCB), induction melting in a cold crucible (IMCC) or self-propagating high-temperature synthesis (SHS).

Stable matrix materials suitable for the HLW conditioning include analogs of long-lived natural minerals based on complex oxides of titanium, zirconium, aluminum and

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Table 1
Approximate number of atoms of some fission products formed in the fission of 1000 uranium atoms in fuel 300 days after the end of the chain reaction [1].

Element	La	Ce	Pr	Nd	Pm	Sm	Y	Zr	Mo	Total
Number of radioactive atoms	–	24	–	–	14	–	1	70	–	109
Total number of atoms	68	149	59	186	14	23	48	320	248	1115

rare-earth elements (REE). The most common RREs are light lanthanides (Ln) with a high level of yield during nuclear fuel fission. Along with other fission products, these can be used to form a conserving matrix for the actinide (An) immobilization. Table 1 presents the content of elements in cooled spent fuel [1] which, together with minor actinides, shape the composition of the An-RRE fraction during the HLW fractionation.

Synthesized analogs of long-lived minerals based on titanates, zirconates and aluminates are the most common matrix materials for the immobilization of An, Ln and other elements of the An-Ln HLW fraction which form mineral-like (ceramic) matrices. Primarily, these are titanate and zirconate pyrochlores (RRE, An, Ca, …)$_2$ (Ti, Zr)$_2$O$_7$ [2,3], titanate and aluminate perovskites (Ca, RRE, An, …) (Ti, Al)O$_3$ [4,5] и Y–Al garnets (Y, Ln, An, Ca, …)$_3$Al$_5$O$_{12}$ [5], which isomorphically incorporate HLW actinides (^{237}Np, ^{239}Pu, ^{241}Am, ^{242}Cm) with the chemical properties and ionic sizes close to those of light Ln.

oxides containing elements of metals with a relatively low affinity to oxygen (Fe_2O_3, MoO_3, MnO_2, CuO and others). The charge also includes structure-forming additives containing oxides of elements which, together with oxides of fuel elements, form the basis for the target SHS product formation. These oxides are largely TiO_2, ZrO_2, Y_2O_3, Al_2O_3, SiO_2, CaO and Ln_2O_3. The elemental composition of the SHS charge depends on the specific target product to be formed, a mineral-like matrix (MLM) suitable for environmentally safe disposal.

Radionuclides of HLW may be added to the SHS charge in the form of HLW calcinates or in the form of sorption calcinates containing radionuclides extracted from liquid HLW, based, e.g., on Al_2O_3 and TiO_2, as it is used in this study with the employment of simulator elements, uranium and americium-241.

The SHS charge formulation is calculated in accordance with the most probable nature of the interaction between the charge components using the yttrium-aluminum garnet formation chemical equation

$$\mathrm{x\ Al} + 0.5\mathrm{x\ Mo0_3} + (2.5 - 0.5\mathrm{x})\underbrace{[(0.102\ \mathrm{CeO_2} \cdot 0.122\mathrm{Nd_2O_3} \cdot 0.224\ \mathrm{ZrO_2} \cdot 0.121\mathrm{MoO_3})\mathrm{Al_2O_3}] + \left(\tfrac{\pi}{2} - \theta\right)}_{\mathrm{SORPTIONCALCINATE}}$$
$$+ 0.1\mathrm{UO_2} + 0.1\mathrm{CaO} + (0.536 + 0.173x)\mathrm{Y_2O_3}$$
$$= \left[\mathrm{Ce}_{0.255-0.051x}\mathrm{Nd}_{0.610-0.122x}\mathrm{Zr}_{0.56-0.122x}\mathrm{Mo}_{0.303-0.061x}\mathrm{U}_{0.1}\mathrm{Ca}_{0.1}\mathrm{Y}_{1.072+0.346x}\right]\mathrm{Al_2O_3} + 0.5x\mathrm{Mo}.$$

The study continues an earlier [6] laboratory research on high-temperature synthesis of matrices based on Y–Al garnet with incorporation of it into the structure of Ce, Nd, Sm, Zr, Mo, U and ^{241}Am. The said elements, considered as model waste, simulate the elements of the An-RRE HLW fraction that are isomorphic to them.

Self-propagating high-temperature synthesis (SHS), a highly technically and economically attractive energy-saving technique, was used to synthesize the matrix at high temperatures with all simulator elements being simultaneously fixed within the structures of its neoformations [7]. The major advantage of the method is that it uses an out-of-furnace technology to implement a high-temperature solid-phase exothermic process in the metal-ceramic SHS mode. In the course of this process, following the priming, e.g. by electrical ignition, spontaneous chemical transformations take place during a short-term heat-up of the reaction mixture to temperatures of 2000–2500 °C in the combustion wave propagating in the initial powder-like or precompacted charge.

The SHS charge formation diagram is shown in Fig. 1.

The metal-ceramic SHS charge includes energy-forming components: fuel – metallic powders of elements with a great affinity to oxygen (Al, Ti, Zr), and oxidizers – oxygen-rich

The experimentally determined value "x" in the equation corresponds to the establishment of the best possible energy conditions of the SHS process.

Apart from the presented chemical equation (option I), two more alike processes were investigated in the study: option II where Sm_20_3 (An simulator) is added to the charge instead of uranium, and option III where ^{241}Am is added to the charge for option I.

For synthesized specimens, the content of the HLW radionuclide simulator elements in the charge was about 10% wt; the content of uranium was about 3% wt; and the specific activity of ^{241}Am in the MLM specimen was $8 \cdot 10^7$ Bq/g.

In option III, the SHS process was implemented in a safety box of the KNZh-2 type accommodated inside a hot laboratory room for activities of radiation hazard class I. The front panel of the box was additionally clad with lead plates, and the windows were covered with lead glass. Inside the box, laboratory equipment was laid out as shown in Fig. 2.

A graphite crucible of the diameter 60 mm with a graphite lid was used as the reaction vessel. The SHS charge was being thoroughly mixed, placed in the crucible, dried at a temperature of 120–130 °C, and pressurized manually. A portion of

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