Encapsulating of high-level radioactive waste with use of ceramic coatings
(Program of investigations)

By

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Introduction

High-level radioactive waste (HLW) which contain approximately 99 % of long-lived fission products and transplutonium elements, constitute a real danger to biosphere.

Applied methods of solidification of HLW with preparation of phosphatic and borosilicate glasses do not guarantee in full measure safety of places of waste-disposal of solidified waste in geological formations during thousand years.

One promising way to improve HLW handling safety is placing of radionuclides in mineral-like matrixes similar to natural materials.

The other possible way to increase safety of HLW disposal places is suggested for investigation by experts of Russian research institutes within the framework of the present program is to introduce an additional barrier on a radionuclides migration path by coating of HLW particles.

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The goals of the Program

The main goals of the Program are:

- Investigations on development of method of HLW encapsulating with use of protective ceramic materials.
- Study of the prepared capsules characteristics.
- Estimation of safety of encapsulated HLW disposal in geological formation.

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Planned researches

To realize the aims the Program will provide the performance of wide circle of researches:

- **Analysis of composition and properties of HLW being subject to solidification and encapsulation**

  In virtue of such analysis the forms (oxides or other stable compounds) and methods of preparation of waste for granulation will be choose.

  Process of preparation of granules of specified types of waste appropriate for protective coatings deposition will be develop.

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Planned researches (continued)

- **Analysis of published and experimental data on properties of coatings.**

  It shall be collected the data on the properties of protective coatings including

  - diffusion coefficients of key fission products;
  - irradiation and corrosion behaviour;
  - strength and thermal strength.

  This will allow to appreciate irradiation and corrosion resistance of capsules for HLW disposal and their mechanical and thermal strength.

- **Designing, manufacture, mounting of lab-scale equipment for preparation of simulators and coating deposition on them.**

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Planned researches (continued)

- **Investigation of coating deposition conditions on HLW simulators.**

  During fulfilment will be
  - manufactured HLW simulators;
  - determined conditions of coatings deposition;
  - developed capsule design requirement;
  - selected and justified capsule design.

  Completion of the task will allow to develop the paperwork of capsule for HLW and process of such capsules preparation.

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Planned researches (continued)

- **Preparation and investigation of characteristics of encapsulated HLW prototypes.**

The fulfilment will allow to test the flow chart of HLW capsules preparation, to manufacture enlarged batches of HLW simulators and experimental batches of capsules, to study their characteristics.

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Planned researches (continued)

- **Development of computation models and computer programs for investigation of processes during storage and disposal of encapsulated HLW. During this task fulfilment will be:**
  - laid down edge conditions of fission products mass transfer through capsule layers including estimation of heat source power in a capsule;
  - estimated fission product release through ceramic coatings at long-term storage.

Solution of this task will allow to predict capsule storage duration, when protective properties will be kept.

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Planned researches (continued)

- **Calculations for selection and substantiation of encapsulated HLW characteristics.**

  Solution of this task will allow to refine initial granules requirements and capsule geometrical dimension, to lay down the final capsule requirements and to estimate their storage duration.

- **Complex analysis of the investigation results and preparation of the initial data for pilot installation design**

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Requirements for protective barriers

The protective barriers must meet the following key requirements:

- low permeability including low coefficients of diffusion of gaseous and solid fission products;
- high radiation and chemical resistance including in ground waters;
- high mechanical strength.

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Materials for investigations

- **Protective barriers:**

PyC
SiC

CVD method:
- low porosity and gas permeability
- low diffusion coefficients

SiC:

$D_{Sr}^{1400C} < 5 \cdot 10^{-17} m^2/s$ **(R.J. Price, 1977)**

$D_{Cs}^{1400C} < 6 \cdot 10^{-21} m^2/s$

PyC: $D_{Cs}^{1200C} \sim 10^{-15} m^2/s$

(D.Stöver, R.Hecker, 1977)

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Protective barriers (continued)

SiC has excellent mechanical, thermal and chemical properties. It is good corrosion resistant material:

Corrosion environment	T,°C	Δl, µm/year	
		Pyrolitic SiC	Reaction-bound sintered SiC (12%Si)
98 % H_2SO_4	100	0.6	18
50% NaOH	100	0.6	<350
53% HF	25	<0.1	2.5
85% H_3PO_4	100	<0.1	3.0
70% HNO_3	100	<0.1	0.2
45% KOH	100	<0.1	<350
25% HCl	70	<0.1	0.3
10% HF+57%HNO_3	25	<0.1	<350

(V.A.Lavrenko et al., 1989)

- **not dissolved in inorganic acids** at the boiling temperatures;
- **concentrated alkalis poorly react with it.**

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Protective barriers (continued)

Oxidation of SiC by oxygen and water vapour begins at temperatures above 400°C.

SIC oxidation rate in dry oxygen is lower than of Si. Addition of moisture increases their oxidation rates. However formed thermodynamically stable SiO_2 film passivates SiC surface and prevents its further oxidation.

Preliminary estimation of the maximum corrosion rate of an ampoule made from the reaction-bound sintered SiC shown that oxide film 0.6 μm in thickness will be formed on its surface at temperature 250°C for one year.

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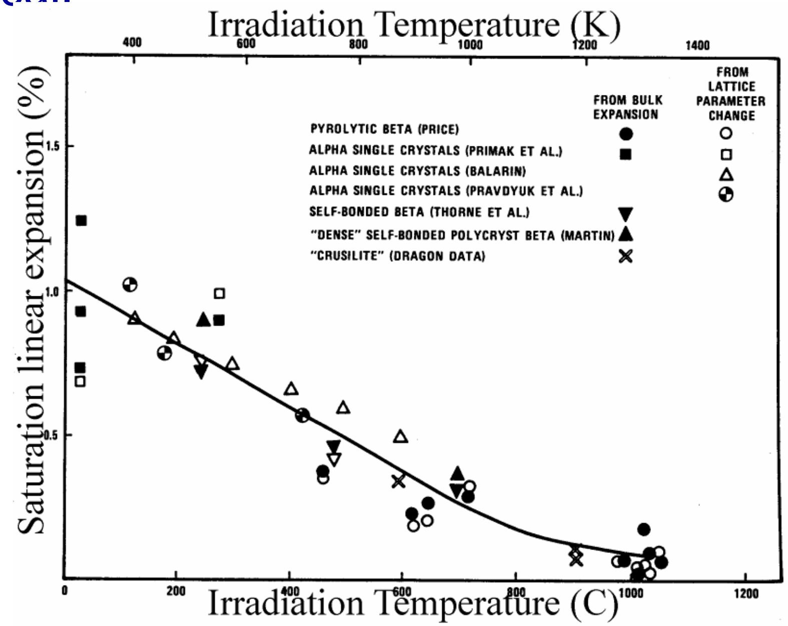
Protective barriers (continued)

Swelling of pyrolitic, reaction-bound sintered and single-crystal SiC at temperatures below 1000°C at least up to fast neutron fluence $5 \cdot 10^{25}$ n/m^2 is practically identical.



All considered factors have predetermined a choice

- **PyC** and **SiC** as protective coatings on granulated waste;
- **SiC** a matrix together with glasses for creation of composites with coated waste granules and/or as a container material

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Materials for investigations

- **Waste**

The following materials will be considered as waste:

- separate fractions of fission products and transuranium elements;
- other products containing long-lived radionuclides (high-level radioactive sorbens formed during purification of liquid waste, calcites).

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Encapsulating process

It is supposed to consider various encapsulating processes of fractionated HLW.

For example

- conversion of HLW solutions into a solid aggregative state (calcites, granules of glass, sorbent, etc.);
- heat treatment of granules;
- deposition of protective coatings;
- placement of coated HLW granules or composites on their base in containers from ceramic material.

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Terms, participants

The Program is calculated for 2.5 – 3 years.

Participation of experts in the fields of high-level radioactive waste processing and their disposal, CVD of ceramic coatings, powder metallurgy, designing and material science from the leading institutes is foreshadowed for carrying-out of the Program.

The cooperation with interested foreign partners is proposed for joint realization of this Program, for example, in the form of an ISTC Project.

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