Immobilization of radioactive vacuum oil into open-porous glass-carbonic material

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Abstract. The paper discusses issues of ensuring safe operation of vacuum facilities in handling radioactive materials. A shaping material for effective immobilization of compound with consolidated organic radioactive waste is proposed.

1. Introduction
Vacuum technologies are widely used in modern science and engineering. Basic tasks set for designing and operation of vacuum systems incorporate reaching high pumping rate of production lines and volumes at the lowest residual pressure. However, problems of personnel and environmental safety are brought to the fore when radioactive material is handled. Minimization of the resultant radioactive waste RW of different types forms the basis for ensuring safety during operation of radioactive vacuum facilities. Since the facilities often use oil vacuum pumps because of their low price, long-term endurance and maintainability, spent vacuum oil contaminated with radioactive components is accumulated in the process of their operation. RW which contain radioactive hydrogen isotope (tritium) present severe problems during storage due to its high migration ability: tritium substitutes non-radioactive hydrogen atoms and is embedded into biological chains as a result of isotopic exchange.

2. Research performed and results obtained
One of the principles enabling to prevent propagation of radionuclides in the environment lies in development of a multi-barrier system of RW immobilization that implies application of two or more independent barriers for waste containment. The barriers are: matrix and buffer materials, casks and waste packages, geological environment of a disposal site, etc. [1]. Consolidated RW in the form of compounds can constitute one of such barriers that provide safety during waste storage and transportation. Requirements to compounds with RW, in particular, include ability of RW compound to endure thermal and mechanical loads, long-term exposure to chemical reagents as well as to withstand dispersion of radionuclide when integrity of other protective barriers (including waste packages) is upset.

The authors earlier proposed a method of consolidation of liquid radioactive waste (LRW) from vacuum oil where a mixture of saturated hydrocarbons is used as a hardening agent [2, 3]. Produced compound reliably immobilizes tritium and its compounds in its structure [4] though has low strength under mechanical loads and thermal resistance.
In order to improve these characteristics in final RW units, the authors deem it reasonable to immobilize waste into a shaping matrix material and foresee extra insulating coating, if appropriate. Compound produced at LRW consolidation serves as the first physical barrier to prevent propagation of radionuclides. This compound should meet legislative requirements related to physical and chemical stability during long-term storage, transportation and final disposal.

Methods of organic LRW immobilization may be divided into two practical directions: (1) adsorption with sorbent followed by encapsulation into matrix material and (2) impregnated immobilization of radionuclides in a preliminary prepared porous matrix through generated differential pressure [5, 6]. Various solids (glue, vermiculite, sand, ground, natural and synthetic fibers, etc.) are used as sorbents. Capillary-porous natural, cement, ceramic or polymer matrices of natural origin and ad hoc are used for impregnation.

Safety principles accepted worldwide and stated in the Russian legislation put forward obligatory compliance with certain public and environmental safety standards in handling sources of ionizing radiation at all stages of life cycle as a basic criterion of RW acceptability [7]. From this viewpoint, LRW immobilization by impregnation of preliminary prepared porous matrices is considered safer because it enables to reduce time of personnel contact with RW, decrease the number of operations, expand the range of conditioned waste and permits to process mixed LRW.

An open-porous glass-carbonic material (OGM) is an advanced matrix material for immobilization of compound with consolidated organic LRW [8]. This material is produced by mixing a binder (liquid resol phenol-formaldehyde resin) and a pore agent (fat solution of oxalic acid in polyhydric alcohol). The pore agent is extracted upon consolidation of the mixture. The resultant workpiece is dried and carbonized in an electric furnace at 850-900°C without air allowed. The process yields a chemically and thermally stable high-porosity carbon-base material which consists of carbon nanoporous microspheres with diameter of 2 up to 50 µm and a developed specific surface. At that, material with different porosity may be produced by varying ratio of primary components. Specimens with porosity of 30 up to 60% were used in our work.

The following criteria were chosen to evaluate efficiency of LRW immobilization: simple impregnation technology, minimum rate of tritium leaching from compound at long-term stay in a contact solution, maximum temperature which rules out dispersion of radionuclide.

Comparative experiments were carried out to optimize an impregnation process, namely: (1) OGM specimens were impregnated with a melt of compound by a contact damping method; and (2) OGM specimens were dipped into the melt for immersion impregnation. At contact damping melt was delivered both through the upper matrix unit plane and through its base. Average index of specimen filling with compound made up 92% from the volume of pore space. Spread of values of a matrix material filling with compound for all specimens did not exceed 1% from the mean value regardless of height of specimens, a melt contact area, total surface area and volume of specimens. Obtained results correlate with data for OGM immersion impregnation by dipping specimens into melt which points to uniform distribution of compound in pore space of the specimens. OGM specimens of different porosity (at least three of each type) were impregnated with a compound simulant to determine thermal stability limit of immobilized compound. The specimens were then heated with a non-contact halogen heater with mass check in a temperature range 90…130° C with step 5° C and 30 min exposure at each step. Free liquid for all specimens started to escape at step temperature transition 120 - 125° C. Taking into account that minimum boiling point of neat vacuum oil is ~140 °C, escape of liquid in this temperature range may be explained by pressure rise inside the specimen driven by light oil fractions or through desorption of gaseous impurities from the surface.

Studies of tritium leaching from the specimens of tritium-containing consolidated vacuum oil were performed as a part of this work. Spent tritium-containing vacuum oil with specific activity of $1.25 \times 10^5$ Bq/g was used to prepare the specimens. Radioactive oil was hardened in mass proportion 50% oil and 50% hardener to produce compound. The compound was used to impregnate a rectangular OGM specimen with sides of 30 mm × 30 mm and height of 5 mm. The specimen was
Enclosed into a hermetically sealed glass container and filled with 50 ml contact solution (distilled water) where it was exposed at 23 ± 2°C with daily water sampling for scintillation analysis.

Tritium was intensively leached within first three days. Then the process almost stopped. Tests were terminated after 15 days since specific activity of contact solution ceased to grow. These data indicate that tritium is leached from the surface and neighboring near-surface layers. Tritium-containing material occupies only some portion of the bulk of a thin layer of immobilized compound. Another portion is occupied by a tritium-inert glass-carbonic material which does not accumulate tritium and participate in its mass transport. A film of radioactive compound was formed on the surface of the specimen at contact damping from which tritium is leached.

The specimen was then cut into pieces, thus, increasing a contact area by 50%. Subsequent scintillation measurements of specific activity of contact solution showed increase of concentration of tritium in it by 45%. This fact is an experimental verification of conclusion on tritium surface leaching.

3. Conclusions
Produced shaping matrix material based on OGM is uniformly impregnated with molten compound in all directions at contact damping under effect of capillary forces which considerably simplifies the technology of impregnation.

Guaranteed thermal stability limit of shaping matrix material with immobilized compound is 110°C. These results meet acceptance criteria placed to radioactive contents and overpacks with consolidated LRW.

New data related to kinetics of tritium leaching from OGM-immobilized compound based on the mixture of saturated hydrocarbons suggests surface phenomena only. Tritium does not migrate from the depth to the surface upon depletion of surface layers and leaching stops.

Immobilization of consolidated radioactive oil into OGM enables to ensure a multi-barrier principle and essentially improve characteristics of resultant RW.

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