
**MATERIALS OF POWER ENGINEERING
AND RADIATION-RESISTANT MATERIALS**

Application of Thermally Expanded Graphite for the Cementation of Cesium- and Tritium-Containing Waste Oils

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Abstract—Thermally expanded graphite (TEG) may be used for solidification of radioactive waste oils produced by the nuclear industry. TEG synthesized within the scope of this work retains the properties of a loose powder-like material when saturated with oil at the ratio of 20 g of oil per 1 g of TEG. At this ratio, the effect of wetting of the cement particles with oil, inhibiting the formation of a monolithic structure, is not manifested. As a result, the required compression strength of a cement compound is retained at the oil content of up to 27 vol %. The effective diffusion coefficients of ¹³⁷Cs and tritium are calculated on the basis of the experimental data on leaching. For the compounds without additives, these diffusion coefficients are $(1.4\text{--}1.6) \times 10^{-13}$ m²/s for both radionuclides. The reason for the coincidence of the diffusion coefficients is the nature of molecular motion inside the winding transport pores of TEG determined by its structure. The diffusion coefficients decrease with time for tritium. The leaching rate of ¹³⁷Cs does not exceed the value specified by GOST for the addition of bentonite to the compound in the amount of 6% by weight of the cement. Application of other mineral and organic additives does not give the desired effect and even increases the leaching rate. The leaching rate of tritium does not depend on additives (including bentonite).

Keywords: radioactive oils, cesium, tritium, cement compounds, thermally expanded graphite, mechanical strength, diffusion coefficient, leaching rate

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INTRODUCTION

During the processing of liquid radioactive wastes (RAW), organic wastes cause a particular problem, most of which are waste oils contaminated with uranium and plutonium isotopes, their fission fragments, tritium, and other radionuclides. This waste as a rule is a mixture of various grades and oils, frequently containing hydrocarbons of lighter fractions, water, and solids. It is clear that such oil cannot be cleaned and reused. Waste oil conversion by oxidation is ineffective owing to the formation of a coke fraction. Therefore, the only method of waste treatment at the moment is temporary storage in metal containers. However, according to the domestic public health regulations, all liquid waste before sending it to long-term storage or burial must be solidified.

Fundamentally, it is possible to use any type of cold formed solid matrices for waste oil solidification. The most readily available suitable material for this purpose is Portland cement. However, a simple cementation has a major drawback: only a small amount of oil (5–7 vol %) may be added to the cement provided that it holds its monolithic shape.

Pre-absorption of oil by porous carbon materials, one of which is thermally expanded graphite (TEG),

makes it possible to achieve a higher degree of oil inclusion in the cement matrix. The result of the oil absorption by TEG is occurrence of a loose product capable of forming composites with mineral binders [1]. An additional advantage of this solidification method is its tolerance to the composition of the organic waste phase.

In accordance with GOST R 51883-2002, cemented radwaste (compounds) must satisfy a number of requirements, the major ones of which are a low leaching rate of radionuclides and high mechanical strength, which is retained under the action of ionizing radiation, after repeated freezing and thawing and a long period of exposure to water. It is well known that the strength and some other characteristics of cement matrixes may be improved by the inclusion of modifying agents.

The purpose of this work is to determine the maximum possible oil content in the TEG-doped cement compound at which the compressive strength still meets the standards (4.9 MPa), to study the ¹³⁷Cs and tritium leaching from such a compound, and to determine the effects of mineral and organic additives introduced into the composition of the compound on the leaching rate.

Table 1. Characteristics of the additives to cement compounds

Additive	Composition	Functions	Content, % by weight of cement
Bentonite	The main mineral—montmorillonite	Moisture permeability reduction	6*
Calcium chloride	CaCl ₂	Solidification catalyst	3.5**
Liquid glass	Na ₂ O · (SiO ₂) _n	Moisture permeability reduction	4****
Melflux	Polyethercarboxylate spray drying products	Plasticizer	0.75*****
S3	Polynaphthalene-methylene-sulfonate sodium salts	Strength increase	0.6*****
D5	Sodium sulfate, andesine, quartz, muscovite, kaolinite	Solidification catalyst, strength increase, frost resistance, moisture permeability reduction.	2.25*****
Nordplast	Solution of complex salts	Plasticizer	0.6*****
		Solidification catalyst, frost resistance	
		Plasticizer	

* Plecas I. and Dimovic S. Influence of Natural Sorbents in Immobilization of Radioactive Waste in Cement. International Conference Nuclear Energy for New Europe 2006 Portoroh, Slovenia, September 18–21, 2006.

** IAEA-tecdoc-1701, The behaviours of cementitious materials in long term storage and disposal of radioactive waste, International atomic energy agency, Vienna, 2013.

*** Korneev V.I. and Danilov V.V. Liquid and Soluble Glass. St. Petersburg: Stroizdat, St. Petersburg, 1996.

**** According to the manufacturer's recommendations.

The research will make it possible to recommend (or not to recommend) the application of modifiers improving the mechanical properties of the cement compound used for solidification of radioactive oils.

EXPERIMENTAL

We used a chromium sulfate synthesis method to produce TEG. This method is economical and can be easily implemented industrially [2]. Foundry graphite GL-1 (GOST 5279-74) was used a raw material. The synthesis comprised the following stages: the graphite surface oxidation with a saturated aqueous solution of commercial chromic anhydride, chemical intercalation of 94% sulfuric acid (c.p.), bisulfate graphite hydrolysis, graphite oxide washing with distilled water, drying at 120°C, and expansion by microwave heating. The bulk density of the obtained TEG was about 10 g/L; the vacuum oil VM-5 content was up to 25 g/g.

The X-ray analysis of the raw graphite was performed with a D2 PHASER diffractometer (Bruker), Cu-K_α radiation, and Ni filter. Electronic photomicrography and \tilde{O} -ray spectrum microanalysis of TEG were carried out using a VEGA 3 LM scanning electron microscope (Tescan) with the INCA Energy+ microanalysis system (Oxford Instruments). Measurement of the specific surface area and the porosity by nitrogen adsorption at 77 K was performed with a QUADRASORB SI automated analyzer (Quantachrome). Adsorption isotherms were treated according to the equation of Brunauer, Emmett, and Teller (BET). To

determine the TEG porosity based on the measurements, the density functional theory was used [3].

Strength tests of the compounds were performed after 28 days of solidification with an R-0.5 mechanical tensile testing machine with the maximum force of 4.9 kN and a 10 ton hydraulic press.

To study the kinetics of leaching of radionuclides, real nuclear wastes were used: VM-1 oil contaminated with ¹³⁷Cs with activity of 2.36 × 10⁴ Bq/kg and VM-5 oil contaminated with tritium with activity of about 10¹³ Bq/kg. In the study of leaching, the latter was diluted with inactive fresh VM-5 oil to 1.2 × 10⁹ Bq/kg. The strength experiments were carried with inactive VM-5 oil.

The cement compounds were prepared by mixing the oil-impregnated TEG (the ratio of the oil mass to the TEG mass being 20) with the cement paste (M-500 cement, the water–cement ratio of 0.3). The list of additives to the cement paste, their nature, basic functions, and the quantities used in the production of the compounds are presented in Table 1. Water-insoluble additives (bentonite, Melflux, S3, D5) were mixed with cement and then charged with mixing water. Soluble additives (CaCl₂, liquid glass, Nordplast) were initially mixed directly with the mixing water.

The obtained cement block represented cylinders with a diameter of 3.6 and a height of 3.5 ± 0.1 cm. The geometric surface area of each block was 61 cm², and the weight was ~47 g. The cement compounds were kept for 28 days for curing and then were placed in closed containers with distilled water. The leaching

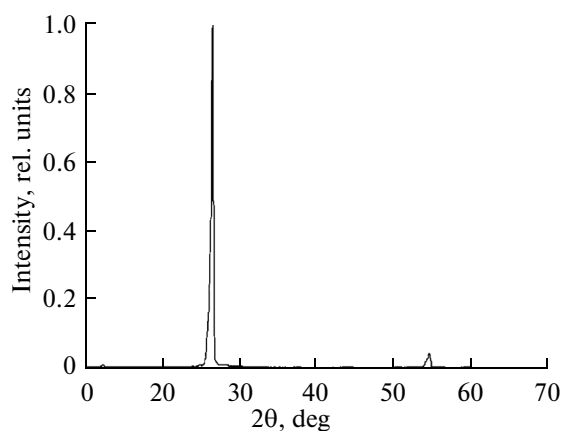


Fig. 1. X-ray diffraction pattern of graphite GL-1.

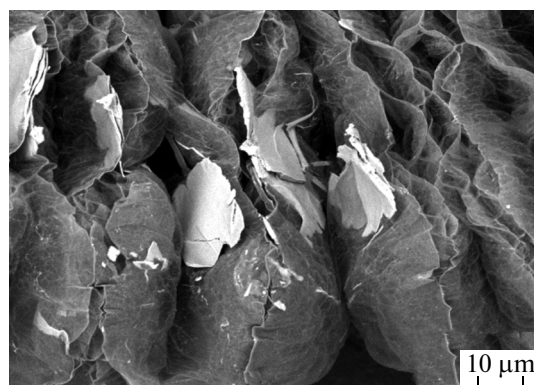


Fig. 2. TEG particle fragment.

was carried out at room temperature without changing the water. A change of the leach water as specified in GOST R 52126 is a standard condition for the testing of glasslike, mineral-like, and ceramic matrixes, i.e., the matrixes for solidification of high-level waste. However, the low radioactivity level of the utilized waste and the low leaching rate lead to the fact that, in view of a periodic water change, the amount of cesium passing into the water is so insignificant every time that the required measurement time increases greatly.

The tritium activity in oil and in leach water was determined by a HIDEK 300SL liquid scintillation spectrometer (Hidex). To measure the activity, we prepared in polyethylene vials aqueous or oil sample solutions (0.5 mL) in 5 mL of Optisafe-2 liquid scintillator. The activity measurement was carried out with a statistical error of not more than 3%, including the sampling error.

The ^{137}Cs activity measurements were performed with an MKS-01A gamma spectrometer (STC Amplituda) with a NaI (Tl) scintillation detector. The leach water left after each sample was poured in full into a separate 1 L Marinelli vessel, and after the measurement, it was brought into contact with the cement compound. The statistical error of the measurements was less than 15%. The base oil activity was defined in the same geometry as the water activity.

The degree of radionuclide leaching degree was calculated by the formula

$$S = \frac{aV}{a_0mv}, \quad (1)$$

Table 2. Elemental Composition (at %) of the TEG surface obtained from graphite GL-1

Area in Fig. 2	Al	Si	Fe	S	Cr	O
Light area	4.5	4.4	1.4	1.1	0.7	8.4
Dark area	0.4	0.7	0.1	1.0	1.2	9.8

where a is the measured radionuclide activity in the sample, Bq; V is the volume of the leach water, mL; a_0 is the oil specific activity Bq/g; m is the oil mass in the cement compound, g; and v is the volume of the water sample, mL.

RESULTS AND DISCUSSION

Figure 1 shows the X-ray diffraction pattern of the raw graphite GL-1. It is possible to see only major reflections typical of the graphite structure: (002) and (004) at 26.56° and 54.65° , respectively [4]. The absence of the reflections (hkl), where $l \neq 0$, indicates that two-dimensional carbon atom networks are arranged relative to each other with a low degree of order. There are no reflections in Fig. 1 with intensity of more than 1% foreign to the graphite structure, which indicates its high chemical purity.

Figure 2 shows an electron photomicrograph of the TEG particle prepared from graphite GL-1. It can be seen that a folded structure with many slit-shaped pores is inherent to TEG. The particles are split mainly toward the atom network stacking and consist of stacks of graphene sheets with a thickness according to the photomicrography no greater than 50 nm. The photograph represents bright areas whose density is significantly greater than the average density of the main phase.

The results of microanalysis of these areas (Table 2) show an increased content of impurities of aluminum, silicon, iron, sulfur, and chromium. The concentration of the first three elements in the areas with a lower density is several times lower. We can conclude that the GL-1 contains aluminosilicate inclusions (which is representative of natural graphite), which are concentrated in individual grains, do not participate in chemical reactions of the TEG synthesis, and do not affect the quality of the final product, i.e., the graphite oxide expansion rate during thermolysis. It should also be noted that all TEG surface segments contain oxy-

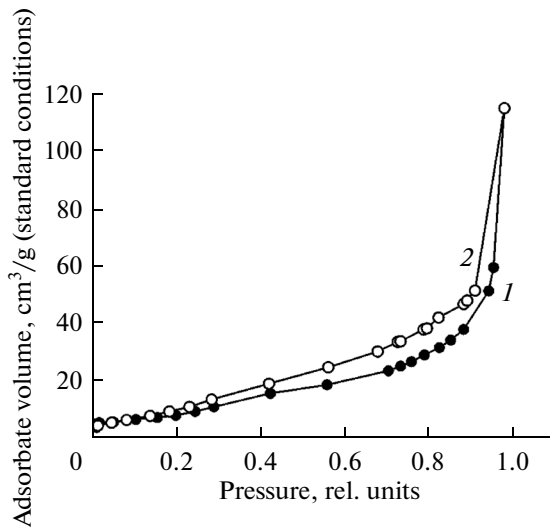


Fig. 3. Nitrogen adsorption isotherm on TEG: (1) adsorption, (2) desorption.

gen—this is typical of all carbon materials. As for sulfur and chromium, the appearance of these elements is caused by the method of synthesis. Their distribution over the TEG surface can be considered more or less even.

A nitrogen adsorption isotherm on TEG (Fig. 3) belongs to the fourth type of BET classification, which is representative of mesoporous solids [5]. The value of the specific surface area is low—only 34 m²/g. The isotherm shows a distinct sorption–desorption hysteresis, indicating the presence of pores with capillary condensation inside them. In fact, as the width distribution analysis of the sorbent pores shows, their size lies within the range of 3 to 15 nm. However, the total volume of these pores is small—less than 0.1 cm³/g. Thus, the low density of TEG is caused by a large volume of macroscopic cavities and broad transport macropores,

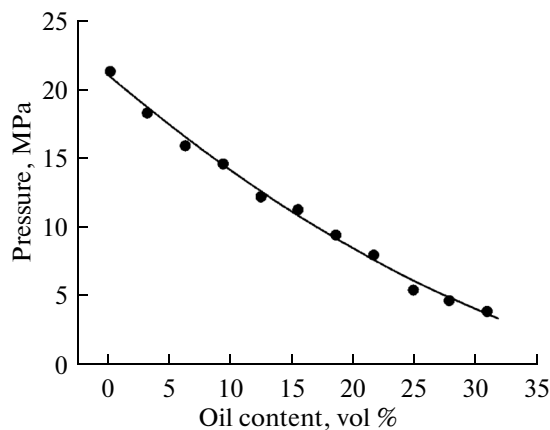


Fig. 4. Dependence of the compressive strength on the oil content in the compound.

which are filled with hydrophobic organic liquids and confine them owing to the capillary effect.

The obtained TEG being saturated with VM-5 oil at a ratio of 20 g/g retains the properties of a loose powder-like material, which makes it possible to produce cement compounds suitable for reliable RAW isolation. At the same time, the TEG holding capacity with respect to oil hydrocarbons is so high that the effect of wetting of the cement particles with oil, preventing hydration of the minerals, is minimal.

Figure 4 shows the dependence of the compressive strength P (MPa) of the obtained cement compounds on the oil content x (vol %). This dependence has the character of a monotonic decrease (in the studied range, it can be described by an empirical dependence $P = 0.0065x^2 - 0.76x + 21$ with a correlation coefficient of 0.994). It follows from this dependence that the required compression strength of the compound is retained at the oil content of up to 27 vol %. Therefore, the experiments on radionuclide leaching were conducted with the compounds containing this maximum oil volume.

Figure 5 shows the kinetic curves of the ¹³⁷Cs leaching using different fillers. Note that a significant release of the radionuclide from the oil during the first few weeks was not expected, so the first data point was obtained only after 4 weeks from the time of the compound contact with water. However, as it turned out, the ¹³⁷Cs yield by this time was significantly higher than the detection limit for all the samples.

Figure 5 allows one to trace different impacts of the cement additives on the degree of ¹³⁷Cs leaching. A positive effect (decrease) is observed on addition of bentonite, which is known as a selective sorbent of cesium and the D5 additive. All other additives give a negative effect. A mathematical treatment of the kinetic curves shows that the best simple equation to

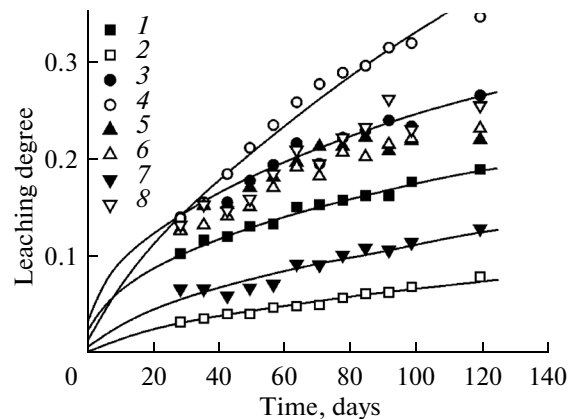


Fig. 5. Kinetic curves of ¹³⁷Cs leaching from the cement compounds without additives (1) and with additives: (2) bentonite, (3) calcium chloride, (4) liquid glass, (5) Melflux, (6) S3, (7) D5, (8) Nordplast.

Table 3. Coefficients of Eq. (2), empirical correlation coefficients, and effective diffusion coefficients of ^{137}Cs for the cement compounds

Additive	A	B	R^2	$D, 10^{-13} \text{ m}^2/\text{s}$
No additives	0.026	0.416	0.988	1.4
Bentonite	0.0046	0.587	0.979	0.18
CaCl_2	0.035	0.424	0.964	2.6
Liquid glass	0.015	0.672	0.978	4.0
Melflux	0.036	0.403	0.913	2.2
S3	0.026	0.471	0.963	2.2
D5	0.0092	0.547	0.867	0.5
Nordplast	0.024	0.503	0.926	2.4

describe them is a power dependence of the leaching degree S on time t (days):

$$S = At^B. \quad (2)$$

The corresponding approximating curves for the compounds with bentonite additives, D5, CaCl_2 , and sodium silicate and for the compound without additives are shown in Fig. 5 with full lines. Table 3 presents the values of the coefficients A and B , as well as empirical correlation coefficients R^2 . As follows from Table 3, the reliability of the approximation of experimental results by Eq. (2) in most cases is sufficiently high (R^2 is not less than 0.9).

Analysis of the parameters of Eq. (2) makes it possible to determine that almost all the samples containing the oil contaminated with ^{137}Cs have representative values of the exponent B in the range between 0.4 and 0.6. This indicates that a rate-limiting step of the cesium leaching is its diffusion in the solid state. Then, using a known solution of the diffusion problem for the conditions of the conducted experiment, we can estimate the effective diffusion coefficient D of ^{137}Cs in the compounds [6].

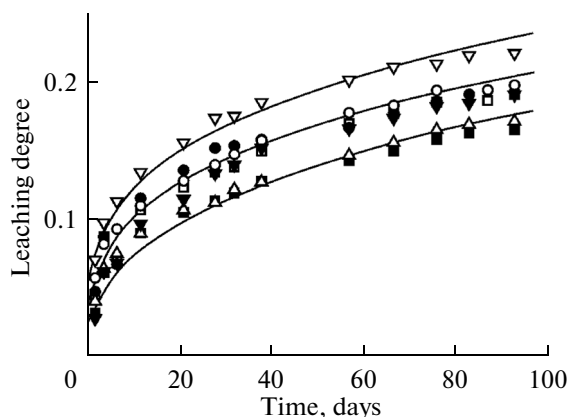


Fig. 6. Kinetic curves of tritium leaching from the cement compounds without additives (1) and with additives: (2) bentonite, (3) calcium chloride, (4) liquid glass, (5) Melflux, (6) S3, (7) D5, (8) Nordplast.

For the desorption of any component from a solid uniform cylindrical particle with a radius R and a height H in a constant volume of fluid (under the condition of a quick balancing of its concentration in the liquid), the relative amount of this component out of the particle by a given time t is

$$S = 1 - \left[\sum_{n=0,1,\dots} \frac{8}{\pi^2(2n+1)^2} \times \exp \left\{ -\frac{4\pi^2(2n+1)^2 Dt}{H^2} \right\} \right] \times \left[\sum_{n=1,2,\dots} \frac{4}{\mu_n^2} \exp \left\{ -\frac{\mu_n^2 Dt}{R^2} \right\} \right], \quad (3)$$

where μ_n are roots of the Bessel function J_0 .

The diffusion coefficients determined from Eq. (3) are presented in Table 3. However, these values are approximate; they make it possible to compare the holding capacity of different compounds directly without extra calculations and particular reference to such parameters as the block size and shape.

In accordance with GOST R 51883-2002, the main value that determines the reliability of radionuclide fixation in the composition of the compound and the chemical resistance of the latter is its leaching rate R_B , determined as

$$R_B = \frac{A'}{A_0 Ft}, \quad (4)$$

where A' is the radionuclide activity (Bq) leached in a given time interval t , days; A_0 is the specific radionuclide activity in the original sample, Bq/g; and F is the area of the open geometric surface of the solid sample, cm^2 . Using (1), we can rewrite the definition (4) as follows:

$$R_B = \frac{Sm}{Ft}, \quad (5)$$

where m is the mass of the solid sample, g. The leaching rates of ^{137}Cs calculated according to formula (5) are presented in Table 4. It should be noted that the leaching rate under the specific value of $10^{-3} \text{ g}/(\text{cm}^2 \text{ day})$ was observed only for the compound containing bentonite.

Kinetic curves for tritium leaching are shown in Fig. 6. We can see that the introduction of additives into the cement paste in this case does not reduce the degree of leaching: the additive S3 hardly changes it, and all others noticeably increase it. As for ^{137}Cs , the kinetic curves may be approximated by Eq. (2); for the compounds containing liquid glass and Nordplast and for the compound without additives, the results of approximation are presented in Fig. 6 with full lines. However, the exponent B in this case is at the level of 0.3–0.4 (Table 5). Consequently, the effective tritium diffusion coefficient in most of the compounds obtained is not constant (otherwise, the exponent

Table 4. Leaching rate of ^{137}Cs from the cement compounds containing TEG, 10^{-3} g/(cm² day)

<i>t</i> , days	No additives	Bentonite	Calcium chloride	Liquid glass	Melflux	S-3	D-5	Nordplast
29	2.83	0.92	3.95	3.79	3.46	3.51	1.83	3.61
36	2.54	0.83	3.55	3.40	3.26	2.92	1.47	3.35
43	2.19	0.76	2.95	3.40	2.82	2.60	1.11	2.70
50	2.04	0.67	2.84	3.32	2.70	2.42	1.10	2.50
57	1.86	0.67	2.73	3.24	2.49	2.38	0.99	2.55
64	1.85	0.62	2.68	3.17	2.42	2.38	1.14	2.59
71	1.70	0.57	2.24	3.06	2.35	2.04	1.02	2.17
78	1.59	0.59	2.25	2.90	2.15	2.09	1.03	2.23
85	1.50	0.58	2.11	2.73	2.05	1.89	1.00	2.15
92–93	1.39	0.54	2.06	2.65	1.77	1.86	0.92	2.20
99	1.40	0.55	1.89	2.53	1.73	1.78	0.92	1.82
120	1.24	0.52	1.75	2.25	1.44	1.52	0.84	1.67

would be equal to 0.5) and decreases with time. This, in turn, may indicate both the variety of tritium compounds in the waste oil [7] and the fact that, upon the compounds being in prolonged contact with water, the chemical formula of the tritium leaching gradually changes.

The effective tritium diffusion coefficients determined from relation (3) are shown in Table 5. Despite the fact that these results are even more approximate than that for ^{137}Cs , the proximity of the diffusion coefficients of tritium and cesium in order of magnitude stands out particularly. In order to give a definite answer whether this is an accidental coincidence or the mechanisms of tritium and cesium diffusion in cement compounds have common limiting stages, we need to perform a separate study. Thus, on one hand, it is necessary to establish the chemical form of the radionuclides in the waste oils; on the other hand, it is necessary to develop the most likely scenario for their transfer in a heterogeneous environment as a combination of elementary steps (adsorption, desorption,

chemical reactions, etc.). Such a task in this paper has not been set.

It should also be noted that the values of the tritium effective diffusion coefficient in the compounds containing TEG are two orders of magnitude lower than in construction concrete (in the form of a bolster) and three orders of magnitude lower than in plain cement brick [8, 9]. The diffusion coefficient of tritium in the waste oil under the same conditions is $D_0 = 5 \times 10^{-11}$ m²/s [10]. It is possible to explain such a low mobility of tritium into the compounds containing TEG by the features of the emerging structure. A highly ordered surface of the TEG graphene sheets determines the long-range order in the adsorption layer of hydrocarbon molecules. The presence of the long-range order and, as a result, the absence of vacancies considerably reduces the mobility of the molecules adsorbed between the graphene sheets. Furthermore, the graphene sheets, not being an absolutely rigid substrate, interacting with hydrocarbon molecules can form a highly labyrinthine pattern. The observed diffusion coefficients in the motion along the winding pores as a rule are much less than in a liquid. The effective diffusion coefficient in this case is defined as

$$D = \frac{D_0 \theta}{\tau},$$

where θ is the fraction of free cross section of the pores, identical to the fractional free volume of the porous body ($\theta < 1$), and τ is the coefficient of tortuosity introduced for the adjustment of the path length and changes in pore diameter ($\tau > 1$) [11]. Thus, for the materials with long winding pores, such as TEG, we should expect a much lower diffusion mobility of the molecules containing tritium than in the oil or in the materials for which tortuous pores are not typical. It should be assumed that precisely the nature of motion of molecules in the winding pores of TEG

Table 5. Coefficients of Eq. (2), empirical correlation coefficients, and effective diffusion coefficients of tritium for the cement compounds

Additive	<i>A</i>	<i>B</i>	<i>R</i> ²	<i>D</i> , 10^{-13} m ² /s
No additives	0.031	0.383	0.958	1.6
Bentonite	0.043	0.340	0.938	2.0
CaCl ₂	0.049	0.319	0.942	2.1
Liquid glass	0.050	0.311	0.991	2.1
S3	0.037	0.347	0.984	1.6
D5	0.028	0.445	0.950	2.0
Nordplast	0.064	0.284	0.989	2.9

Table 6. Leaching rate of tritium from the cement compounds containing TEG, 10^{-3} g/(cm² day)

<i>t</i> , days	No additives	Bentonite	Calcium chloride	Liquid glas	S-3	D-5	Nordplast
2	12.2	16.4	18.5	22.1	15.8	11.1	27.9
4	12.0	16.4	17.4	16.0	12.7	12.0	19.0
7	7.62	10.4	10.6	10.5	8.48	9.56	12.5
12	5.78	6.84	7.53	7.12	5.84	6.24	8.67
21	3.90	4.53	5.02	4.68	3.95	4.28	5.74
28	3.19	3.75	4.22	3.87	3.16	3.70	4.80
32	2.91	3.32	3.73	3.56	2.96	3.39	4.25
38	2.60	3.03	3.28	3.20	2.63	3.10	3.78
57	1.95	2.32	2.40	2.43	2.01	2.26	2.74
67	1.73	2.05	2.08	2.13	1.81	2.01	2.45
76	1.62	1.90	1.92	1.98	1.70	1.87	2.18
86	1.52	1.70	1.79	1.77	1.58	1.72	2.05
93	1.38	1.61	1.61	1.65	1.43	1.59	1.84

leads to the coincidence of the effective diffusion coefficients for tritium and cesium.

Despite the fact that there is no standard tritium waste leaching rate, we have defined this value according to (5), as in the case of ¹³⁷Cs. The results of evaluation are presented in Table 6.

As expected, the leaching rate of tritium in the first few days significantly (factors of ten) exceeds the standard for ¹³⁷Cs. However, in view of the differences in the radiation energy of these radionuclides, we can conclude that the environmental damage in the case of contact with tritium from oil-containing compounds will be significantly smaller. In addition, the studies show [12] that the covering of cement compounds with hydraulic insulating materials (e.g. epoxy resin) is able to reduce the leaching rate of tritium by more than an order of magnitude, reducing it to the standard applied to ¹³⁷Cs.

CONCLUSIONS

TEG, which preserves properties of a loose powder material when saturated with VM-5 oil at a ratio of 20 g/g, has been synthesized by the chromium sulfate method from graphite GL-1. At this ratio of oil and TEG, the wetting of the cement particles with oil preventing the formation of a monolithic structure it not yet manifested. This makes it possible to obtain oil-containing cement compounds suitable for the safe isolation of radioactive waste. It is found that the required strength of the cement compound for compression (4.9 MPa) is retained at the oil content of up to 27 vol %.

The studies of the chemical stability of oil-containing compounds of different compositions indicate that

the rate-limiting leaching step of the radionuclides is their diffusion in the solid state. The effective diffusion coefficients of ¹³⁷Cs and tritium were calculated on an experimental basis, making it possible to compare the holding ability of different compounds directly, without additional calculations and without reference to the size and geometry of the unit. The diffusion coefficients in the compounds without additives are $(1.4-1.6) \times 10^{-13}$ m²/s both for ¹³⁷Cs and for tritium. The most likely cause of such a coincidence is the nature of motion of molecules inside the TEG winding transport pores, determined by its structure. The diffusion coefficients for tritium decrease with time. This may be due to the fact that the waste oils contain tritium in various chemical forms; thus, transitions between the various forms of tritium cannot be excluded from the formation of the compound or during leaching.

The ¹³⁷Cs leaching rate does not exceed the standard value of 10^{-3} g/(cm² day) only when bentonite is added to the compound (a cesium selective sorbent) in the amount of 6% by weight of the cement. Introduction of other mineral and organic additives applied in the practice of concrete production for improving the strength properties, in most cases, increases the leaching rate of cesium (an exception is the D-5 modifier). Thus, their application in the cementation of oils containing cesium is impractical. The introduction of additives (including bentonite) has practically no effect on the leaching rate of tritium. The leaching rate of tritium can be reduced by a waterproof coating of the cement blocks.

It is shown that thermally expanded graphite may be used for solidification of radioactive waste oils produced in the nuclear industry.

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