



Article

Leaching Behavior of Cs-137 from Cement-Based Matrices for Immobilization of Irradiated Uranium–Graphite Fuel

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Abstract

During the operation of the first core of the IGR pulsed graphite reactor (1961–1966), 2.6 tonnes of irradiated highly enriched uranium (HEU)–graphite fuel were accumulated. For its immobilization, cementation with preliminary down-blending using depleted uranium was proposed, with the ²³⁵U concentration in the final matrix limited to less than 50 ppm. Among the radionuclides accumulated in irradiated uranium–graphite fuel, ¹³⁷Cs is of particular concern because it is a long-lived uranium fission product with high mobility in cementitious systems. In this study, four cement compositions with water-to-binder ratios of 0.38–0.42 and Portland-cement-to-fly-ash ratios of 1:2.5 and 1:1.77 were investigated for the immobilization of irradiated uranium–graphite fuel. The ¹³⁷Cs activity in the cement samples was determined using the substitution method with a point calibration gamma source and the MCNP5 code to calculate correction factors accounting for the measurement geometry and gamma-ray self-absorption. For all compositions, the initial ¹³⁷Cs leaching rates exceeded 1×10^{-3} g/(cm²·day) during the first 1–3 days of testing; however, from day 7 onward, the leaching rates decreased below the regulatory limit and continued to decline during the subsequent test period. Based on these results, as well as on the leachability of calcium and silicon dioxide, two candidate cement–matrix compositions were selected. The results provide an experimental basis for further scale-up studies aimed at the immobilization of irradiated HEU graphite fuel from the IGR reactor.

Keywords: Cs-137; leaching; immobilization; cement-based matrices; uranium–graphite fuel



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1. Introduction

The IGR pulsed graphite reactor (Institute of Atomic Energy, National Nuclear Center of Republic of Kazakhstan, NNC RK) has been operated since 1960 and is characterized by a high thermal neutron flux density (7×10^{16} n/cm²·s) and high prompt power; it is intended for testing reactor fuel under extreme conditions [1,2]. During the operation of the first reactor core from 1961 to 1966, 2.6 tonnes of irradiated highly enriched uranium (HEU)–graphite fuel were accumulated. The uranium enrichment by ²³⁵U in the fuel is 90%, the fuel burnup is less than 1%, and the neutron fluence is 4×10^{18} n/cm².

Previously, the immobilization of HEU fuel had not been carried out in Kazakhstan; therefore, it was necessary to develop a dedicated technology, including the implementation of the process line [3]. Taking into account international experience and the results of R&D, cementation was chosen as a widely used method in the management and disposal of low- and intermediate-level radioactive waste [4]. Before cementation, it is proposed to reduce the fuel enrichment by mixing irradiated graphite fuel with depleted uranium powder, followed by encapsulation in Portland cement. Fly ash (FA) with different ratios to Portland cement (PC) was considered as a supplementary additive [5,6]. The mass concentration of ^{235}U to the matrix is maintained at 50 g/tonne, which is close to the natural concentration and makes it possible to transfer the nuclear material to the radioactive waste category. Based on calculations of specific activity, the proposed matrices will be classified as intermediate-level radioactive waste.

After down-blending and the immobilization of the irradiated HEU fuel, the waste form is planned to be placed in 208 L drums. Each drum will contain a cement matrix with a volume of approximately 200 L and a total component mass of about 305 kg, including two IGR core fuel blocks. The drum material is corrosion-resistant steel with a wall thickness of approximately 1 mm. The calculated specific activities of beta-emitting, alpha-emitting, and transuranic radionuclides in the cement matrix containing IGR fuel are summarized in Table 1.

Table 1. Calculated specific activities of beta-emitting, alpha-emitting, and transuranic radionuclides in the cement matrix containing IGR fuel.

Radionuclide Group	Specific Activity, kBq/kg
Beta-emitting radionuclides	2.1×10^3
Alpha-emitting radionuclides, excluding transuranic radionuclides	9.9
Transuranic radionuclides	4.4×10^{-1}

Among the long-lived radionuclides accumulated in irradiated uranium–graphite fuel after approximately 60 years of storage, ^{137}Cs and ^{90}Sr are of particular concern for cement-based immobilization. ^{137}Cs is the predominant gamma-emitting radionuclide in the IGR reactor fuel. Due to the relatively high mobility of cesium in cementitious systems, the ^{137}Cs leaching rate is one of the key performance indicators for assessing the suitability of cement matrices for radioactive waste immobilization [7–9]. ^{90}Sr is a beta-emitting radionuclide and its determination in leachate solutions requires a separate radiochemical separation and beta-counting procedure, which was outside the scope of the present study.

The influence of various additives on cement hydration, as well as ways to reduce radionuclide leaching from a cement matrix, are well described in the overview in [5]. For example, the authors of [10,11] concluded that the addition of natural zeolites reduced cesium release by 70–75% of the amount initially bound in the resin over a 3-year leaching period. The addition of kaolin clay, with kaolin content of 5% in cement, also helps to improve radionuclide retention; however, increasing the additive concentration leads to a loss of mechanical strength and hydrolytic stability. Magnetite, used as an iron-oxide ion exchanger for radionuclides, makes it possible to reduce the leachability of radionuclides [12]. The authors of [13] developed a method for immobilizing technetium in a cement matrix by adding sodium diethyldithiocarbamate at a concentration of 1 wt.%. The addition of a relatively small amount of additive reduced technetium loss from the matrix material by 90%, while maintaining its volumetric stability and mechanical properties.

For IGR fuel, the addition of fly ash makes it possible to obtain a homogeneous matrix structure and improve the performance properties of the matrix (mechanical strength, frost resistance, resistance to prolonged exposure to water, radiation, and leaching) [14].

According to earlier studies [15], four cement–matrix compositions with fly ash additive were selected that met the waste acceptance criteria (WAC), including requirements for the technical and performance (physicochemical) characteristics of waste matrices for the immobilization of radioactive waste. The technical criteria cover parameters ensured at the manufacturing stage (radiation safety, limits on ^{235}U and graphite content, size and uniformity of fuel particle distribution, bleed water ≤ 3 vol.%). The performance criteria regulated by state standards include strength of at least 10 MPa, resistance to freeze–thaw cycling and water immersion (strength loss $\leq 25\%$), and a ^{137}Cs leach rate not exceeding 10^{-3} g/(cm²·day).

This paper describes the methodology and presents the results of leachability tests of ^{137}Cs and matrix constituents, including Ca and SiO_2 , together with measurements of the pH and electrical conductivity in the leachate solutions. Several cementitious matrices based on fly ash and Portland cement were investigated as candidates for the immobilization of irradiated HEU graphite fuel from the IGR reactor. In this study, radionuclide leachability is understood as the ability of a cement compound to retain radionuclides under prolonged exposure to aqueous solutions.

Leaching results obtained over 1 to 29 days make it possible to assess changes in the chemical stability of cement compounds under standardized aqueous leaching conditions and to evaluate their suitability for the immobilization of uranium–graphite fuel from the IGR reactor.

2. Materials and Methods

To determine the radionuclide leaching rate, cylindrical samples of four compositions containing irradiated HEU fuel were prepared (Figure 1); they differed in the water-to-binder ratio, in the range of 0.38 to 0.42, and in the Portland cement/fly ash ratio (Table 2). The compositions were selected based on preliminary tests of a cement matrix with a fuel simulant for bleed water, compressive strength, frost resistance, and water resistance [15,16].

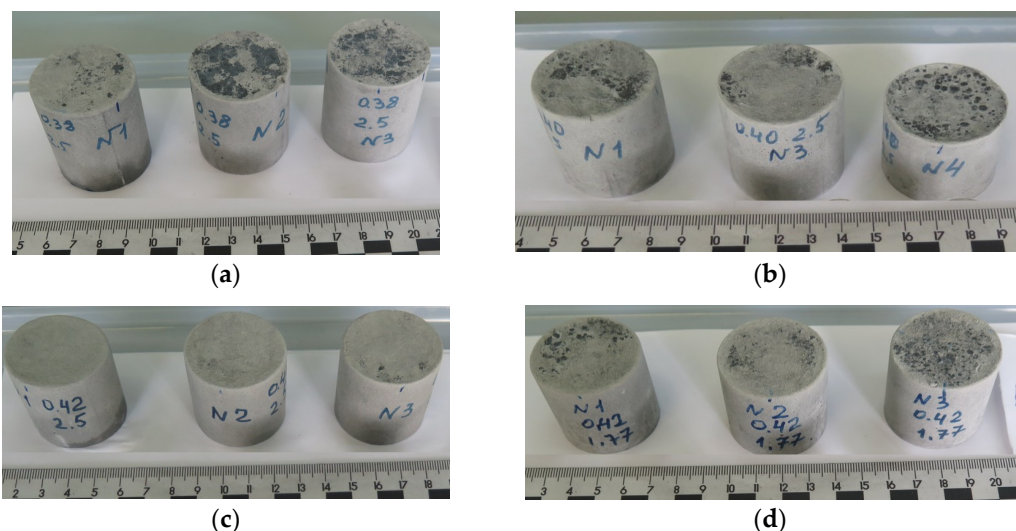


Figure 1. Samples' appearances after 28 days of curing. (a) Composition A: W/B = 0.38, FA/PC = 2.5; (b) Composition B: W/B = 0.40, FA/PC = 2.5; (c) Composition C: W/B = 0.42, FA/PC = 2.5; (d) Composition D: W/B = 0.42, FA/PC = 1.77.

The following materials were used to prepare the cement compound samples:

- Ground fragments of irradiated HEU fuel blocks from the first core of the IGR reactor. The fuel blocks are composed of 11-03-grade graphite impregnated with uranyl dinitrate $\text{UO}_2(\text{NO}_3)_2$. The average uranium concentration in the graphite throughout

the reactor core is 2.7 g/kg; the uranium enrichment is 90% by isotope ^{235}U . The mass fraction of fuel in the cement compound samples was 1.68%. The particle size of the ground fuel fragments did not exceed 200 μm .

- Portland cement of grade CEM I 42.5N (trade name “Super 500”, manufactured by PC Cement Plant Semey LLP, Semey City, Kazakhstan). Most Portland cement particles (97.1%) were less than 100 μm in size.
- Fly ash from thermal power plants for concrete, coal-derived, grade II, acidic type, category B (manufacturer: BEST BUSINESS LLP, Karaganda City, Kazakhstan). Particle size was less than 100 μm .

Table 2. Matrix compositions for determination of the leaching rate.

Composition	Ratios by Mass	
	W/B [Water/(Cement + Fly Ash)]	PC/FA [Portland Cement/Fly Ash]
A	0.38	1/2.5
B	0.40	1/2.5
C	0.42	1/2.5
D	0.42	1/1.77

Table 3 presents the results of the analysis of the compositions of Portland cement and fly ash.

Table 3. EDS data for Portland cement and fly ash.

Oxides	Content, % by Weight								
	Na ₂ O	MgO	Al ₂ O ₃	SiO ₂	SO ₃	K ₂ O	CaO	TiO ₂	Fe ₃ O ₄
Portland cement	0.70 ± 0.05	1.42 ± 0.06	4.68 ± 0.14	16.7 ± 0.3	8.1 ± 0.5	1.28 ± 0.06	62.0 ± 0.4	-	5.1 ± 0.2
Fly ash	0.54 ± 0.11	0.52 ± 0.07	30.9 ± 0.3	62.1 ± 1.0	-	0.85 ± 0.09	2.4 ± 0.2	1.4 ± 0.12	1.16 ± 0.15

“-” means: not defined.

The cement compound samples were prepared according to the following procedure:

- (1) The required amount of irradiated HEU fuel was ground to a particle size of less than 200 μm .
- (2) For each composition, a single batch of cement paste was prepared. The components were added sequentially in the following order: water, fly ash, ground HEU fuel, and Portland cement. The mixture was manually mixed with high intensity for 8–10 min to promote the uniform distribution of the components.
- (3) The prepared cement paste was placed into calibrated cylindrical molds composed of PVC pipes. Before filling, the inner surfaces of the molds were coated with a thin layer of release agent that did not stain the sample surfaces or affect the properties of the surface layer of the cement compound. To improve the homogeneity of waste distribution between parallel samples of the same composition, the molds were not filled sequentially one after another. Instead, the cement paste was distributed among all molds alternately in small portions until they were completely filled. The molds were filled flush with the upper edge, and excess paste was removed. The cement paste was compacted in the molds using a sieve shaker with a vibration amplitude of 0.5 mm for 10 s; the compaction procedure was repeated twice.
- (4) The molds containing the cement paste were placed on a stand inside a closed container with water; the molds did not come into direct contact with water. The curing temperature was 22 °C. The samples were removed from the molds 4 days after preparation. After demolding, the samples were visually inspected, their geometrical

dimensions were measured, and their density was determined. The samples were then returned to normal curing conditions until they reached an age of 28 days.

Depleted uranium was not included in the laboratory specimens, as it is used solely for fuel down-blending in the full-scale immobilization technology and was not expected to influence the ^{137}Cs leaching behavior investigated in this study.

For each composition, three cylindrical samples were produced, with an open surface area of $98.0 \pm 7.2 \text{ cm}^2$. The dark spots and surface heterogeneities visible in Figure 1 are associated with the dispersed local fly ash-rich areas and surface porosity formed during curing. Visual inspection did not reveal the macroscopic segregation of the mixture or local concentration of the waste component in separate zones of the samples.

The geometrical parameters, mass, and density of the samples are given in Table 4.

Table 4. Geometrical and physical parameters of samples.

Composition	Sample Number	Diameter, mm	Height, mm	Mass, g	Volume, cm^3	Density, g/cm^3
A	1	46.13	50.5	142.88	84.39	1.69
	2	46.12	50.5	138.66	84.38	1.64
	3	46.18	51.0	143.53	85.42	1.68
B	1	46.10	43.0	118.27	71.78	1.65
	2	46.13	42.0	116.43	70.20	1.66
	3	46.16	44.5	123.13	74.47	1.65
C	1	46.16	42.5	119.46	71.1	1.68
	2	46.07	44.5	123.76	74.2	1.67
	3	46.14	45.0	124.50	75.2	1.65
D	1	46.14	40.8	115.30	68.1	1.69
	2	46.06	42.4	119.55	70.6	1.69
	3	46.10	41.3	117.86	68.9	1.71

Leachability tests of the cement compounds were carried out after sample curing for 28 days under normal hardening conditions, in accordance with a methodology based on regulatory documents ST RK 3723-2021, Cemented Radioactive Waste, and General technical requirements GOST 29114-91, Radioactive Waste, as well as the method for measuring the chemical durability of solidified radioactive waste by prolonged leaching: GOST R 52126-2003, Radioactive Waste. The determination of the chemical durability of solidified high-level waste was achieved via the prolonged leaching method.

In accordance with these requirements, the most representative components of the matrix and the radiation source, namely Ca and ^{137}Cs , respectively, could be analyzed. The leaching experiments included the following stages:

- (1) Sample inspection for external defects, weighing, and measurement of dimensions;
- (2) Determination of ^{137}Cs activity in the samples;
- (3) Placement of each sample in a plastic container containing the contact solution (300 mL of distilled water);
- (4) Replacement of the spent contact solution after specified time intervals with fresh solution;
- (5) Examination of the spent contact solution: activity analysis and measurement of pH, electrical conductivity, and Ca^{2+} and SiO_2 concentrations.

2.1. Determination of ^{137}Cs Activity in the Samples

The ^{137}Cs leaching rate R_n^i ($\text{g}/(\text{cm}^2 \cdot \text{day})$) was calculated according to GOST 29114-91 and GOST R 52126-2003 as a value normalized to the open surface area and the leaching interval duration via the formula

$$R_n^i = \frac{a_n^i}{A_0^i \cdot S \cdot \Delta t_n}, \quad (1)$$

where a_n^i is the activity of radionuclide i leached to the contact solution during the n -th leaching interval t_n (days), Bq.

A_0^i —specific activity of the radionuclide i in the sample, Bq/g;

S —open surface area of the sample, cm²;

Δt_n —duration of the n -th leaching interval, days, calculated as

$$\Delta t_n = t_n - t_{n-1} \quad (2)$$

t_n —cumulative leaching time from the beginning of the test, $t_n = 1, 3, 7, 10, 14, 21, 29$ days.

Figure 2 shows the gamma-ray spectrum of one of the samples, measured with a CANBERRA InSpector-2000 gamma spectrometer (Mirion Technologies, Atlanta, GA, USA) equipped with a 3'' × 3'' NaI(Tl) scintillation detector. The predominant radionuclide in the spectra is ¹³⁷Cs. The gamma-ray peaks attributed to ¹³⁷Cs are highlighted in blue.

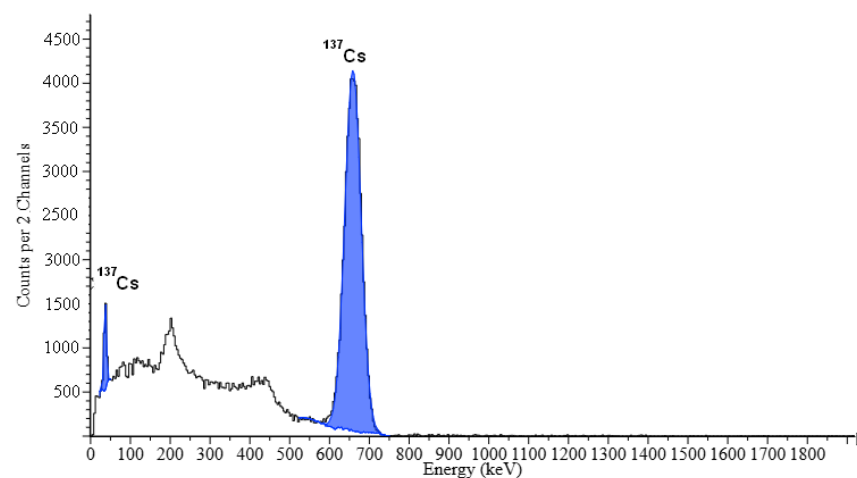


Figure 2. Gamma-ray spectrum of a cement matrix sample.

The activity of the cement matrix samples was determined using the substitution method [17]. This method is applicable when calibration standards with a density and geometry similar to those of the measured samples are unavailable. In this case, the analytical expression for activity determination (2) uses the gamma-ray detection efficiency of a point ¹³⁷Cs reference source, while a correction factor is introduced to account for differences in density and geometry between the point source and the sample.

The measurement geometry was identical for both the point reference source and the samples: the distance between the measured object and the detector cap was 150 mm; the spectrum acquisition time was set to obtain more than 8×10^4 counts in the 661.6 keV full energy peak (FEP). Following the sample measurements, a background spectrum was acquired.

The activity of ¹³⁷Cs in a sample is determined according to the equation

$$A_{sam} = \frac{A_{ref_0}}{D} \frac{\bar{I}_{sam}}{\bar{I}_{ref}} \cdot k, \quad (3)$$

where the symbols *sam* and *ref* refer to the sample and the reference point ¹³⁷Cs source, respectively.

A_{sam} , A_{ref_0} —activity of the sample at the time of measurement and activity of the reference source at the time of certification, Bq;

$\bar{I}$ —average gamma radiation intensity from six measurements, counts;

D —correction for decay of ^{137}Cs , r.u.;

k —correction accounting for the difference in geometry and the self-absorption of gamma rays in the sample and the point reference source [18].

In this study, correction k was calculated via the MCNP5 simulation code using the F8 tally. The correction factor k was calculated by the formula

$$k = \frac{\varepsilon_{ref}}{\varepsilon_{sample}}, \quad (4)$$

where ε is the detection efficiency in FEP for 661.6 keV gamma rays for a point non-self-absorbing gamma source and the sample for an identical measurement geometry.

2.2. Determination of ^{137}Cs Activity in the Spent Contact Solution

The activity of the aqueous solutions obtained during the leaching of the cement matrix samples was determined using calibration based on a volumetric ^{137}Cs standard solution source. The spent contact solution volume was approximately 298 mL (± 2 mL). Before measurement, the solutions were filtered and poured into a measuring container. The filters were measured separately, and the ^{137}Cs activity retained on them was negligible. The volume of the filtered leachate solution in the measurement container was approximately 200 mL (± 0.5 mL). The container was placed directly on the detector endcap.

The solution spectra were measured repeatedly. The spectrum acquisition time was adjusted to ensure that the area of the 661.6 keV FEP contained at least 1×10^3 counts. Prior to FEP area determination, the background spectrum was subtracted from the measured gamma-ray spectrum of the solution.

2.3. Determination of pH, Electrical Conductivity, and the Concentrations of Ca^{2+} and SiO_2 in the Spent Contact Solution

The leaching rate of the cement matrix components was assessed from changes in pH, electrical conductivity, and the concentrations of Ca^{2+} and SiO_2 in the spent contact solutions. Changes in pH characterize the buffer properties of the system and the degree of calcium hydroxide leaching. Electrical conductivity reflects the total concentration of dissolved ionic species in the leachate. The Ca^{2+} concentration serves as an indicator of the decalcification of the cement matrix, whereas the SiO_2 content may indicate the degradation of the C-S-H phase.

These parameters were determined using the following equipment:

- Mark-603 conductivity meter No. 154 (LLC VZOR, Nizhny Novgorod, Russia), for measuring the specific electrical conductivity of distilled water;
- Seven2Go pH meter, model S2 (METTLER TOLEDO, Greifensee, Switzerland);
- KFK-3-01 photoelectric photometer (Zagorsk Optical and Mechanical Plant, Sergiev Posad, Russia);
- Analytical balance with a measurement range of 0.001–200 g and a weighing error of 0.1 mg.

3. Results and Discussion

3.1. Results of Determining the Activity of the Cement Matrix Samples

The correction k accounting for the difference in geometry and the self-absorption of gamma rays in the samples and in the reference source was calculated by MCNP5 with the DLC-220 photon constants library in a geometry reproducing the measurement conditions. The number of particle histories was 2 million, and the statistical error was less than 0.5%. The detection efficiency for a point non-self-absorbing source, ε_{point} , was 5.182×10^{-3} .

The detection efficiency for the samples depends on their composition, density, and height. Given the more significant influence of the latter two, the elemental composition of the samples in the MCNP model was taken to be the same (Table 5) and was not changed for the whole calculation series.

Table 5. Sample elemental composition in the MCNP model for compositions A, B, C, and D.

Element	O	Na	Mg	Al	Si	S	P	K	Ca	Ti	Fe	H	C
Content, %	57.40	0.27	0.31	7.20	13.48	0.67	0.03	0.51	9.78	0.34	1.10	3.71	5.23

The dependence of the correction factor k on the sample density and height was derived from a linear approximation of a two-variable function. The density effect was assessed at the mean sample height, whereas the height effect was assessed at the mean sample density. The results are presented in Figure 3.

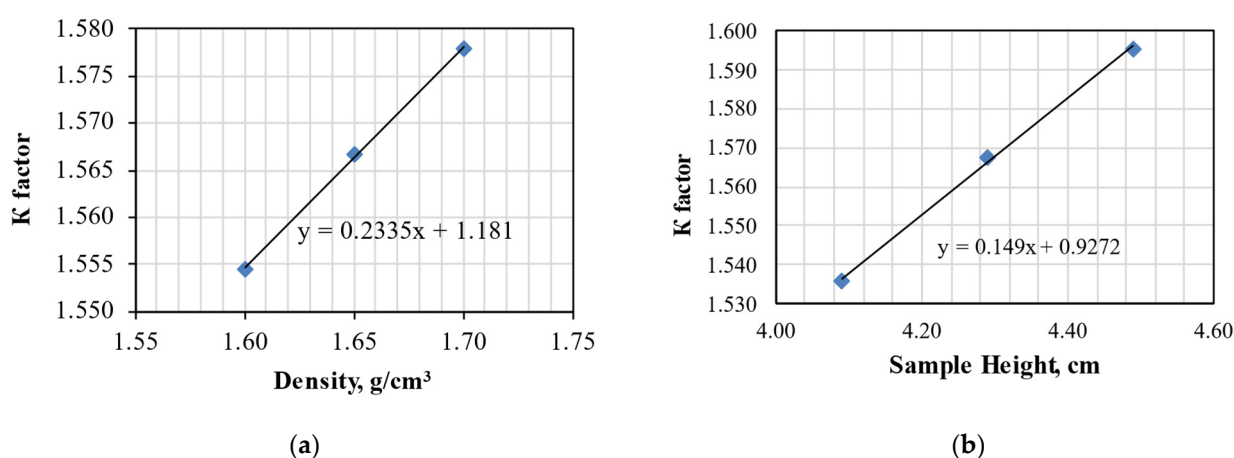


Figure 3. Dependence of the correction factor k on the sample density (a) and height (b).

As a result, the analytical dependence of the correction factor was obtained as a function of two variables: sample density ρ and height h .

$$k = 0.2335\rho + 1.181 + 0.1490(h - h_{aver}) \quad (5)$$

Table 6 presents the activity and specific activity of the cement matrix samples, averaged over three samples for each composition.

Table 6. Mean activity of the cement matrix sample compositions.

Composition	A , Bq	A_{spec} , Bq/g
A	$(1.32 \pm 0.09) \times 10^5$	$(9.32 \pm 0.70) \times 10^2$
B	$(1.17 \pm 0.09) \times 10^5$	$(9.80 \pm 0.70) \times 10^2$
C	$(1.22 \pm 0.09) \times 10^5$	$(9.96 \pm 0.70) \times 10^2$
D	$(1.17 \pm 0.09) \times 10^5$	$(9.95 \pm 0.70) \times 10^2$

3.2. Results of Determining the ^{137}Cs Leaching Rate

After immersion in the contact solution, the samples underwent water saturation, which was accompanied by an increase in mass (Figure 4). The most significant mass gain (approximately 4 wt.%) occurred during the first two days of exposure. Thereafter, the increase in mass recorded at subsequent measurement intervals did not exceed 1 wt.%. After 28 days of leaching, the total mass gain reached approximately 4.8 wt.% for

compositions A–C and 4.3 wt.% for composition D. This difference is likely attributable to the varying cement content of the compositions.

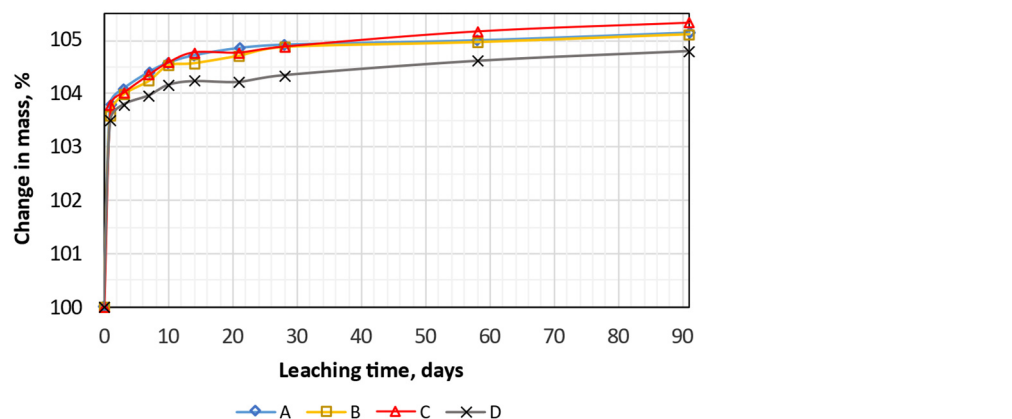


Figure 4. Increase in mass for samples of different compositions during leaching tests.

The dynamics of the changes in ^{137}Cs activity in the contact solutions are presented in Figure 5. It can be seen from Figure 5 that compositions B and D retain the alkali radionuclide better. The relative error in determining the ^{137}Cs activity in the solution does not exceed 6%.

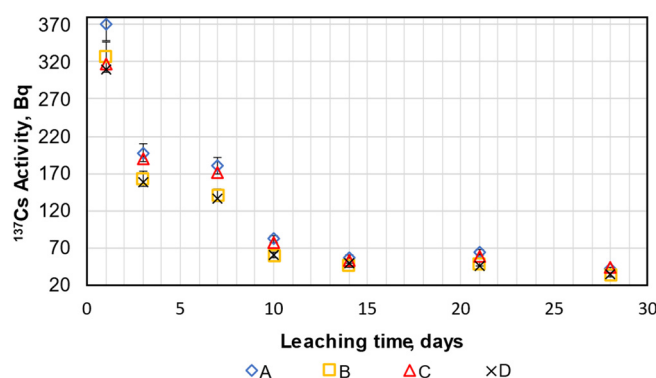


Figure 5. Changes in ^{137}Cs activity in the contact solution.

The ^{137}Cs leaching rates from the cement matrix samples for the A, B, C, and D compositions are shown in Figure 6.

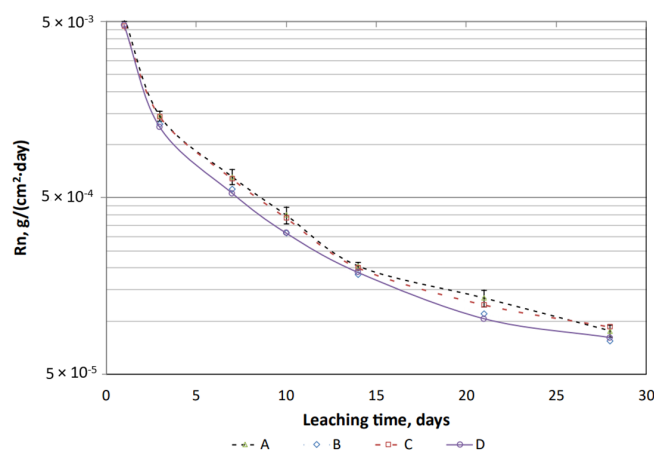


Figure 6. Changes in the ^{137}Cs leaching rate in the contact solutions.

Within the measurement uncertainty, the ^{137}Cs leaching rate values were practically identical throughout the entire test period for cement matrices of all compositions. On days 1 and 3, the leaching rates exceeded the limiting value of 1×10^{-3} specified in ST RK 3723-2021 for cement compound quality requirements. The leaching rates reached acceptable levels by day 7 and continued to decrease thereafter. The high initial leaching rate is attributed to the dissolution of salts on the block surface, whereas its subsequent decrease and stabilization are governed by the diffusion transport of radionuclides from the matrix interior to its surface.

3.3. pH and Electrical Conductivity of the Spent Contact Solution

All investigated compositions showed a common pattern of changes in pH and electrical conductivity (Figure 7). The correlated behavior of these parameters (Figure 7b) with ^{137}Cs activity is reasonably explained by the influence of the concentration of the most mobile free charge carriers OH^- . The increase in pH, accompanied by a simultaneous decrease in electrical conductivity during the last test period, is nevertheless unusual. However, no further increase in pH was recorded when the leaching duration was extended to 90 days.

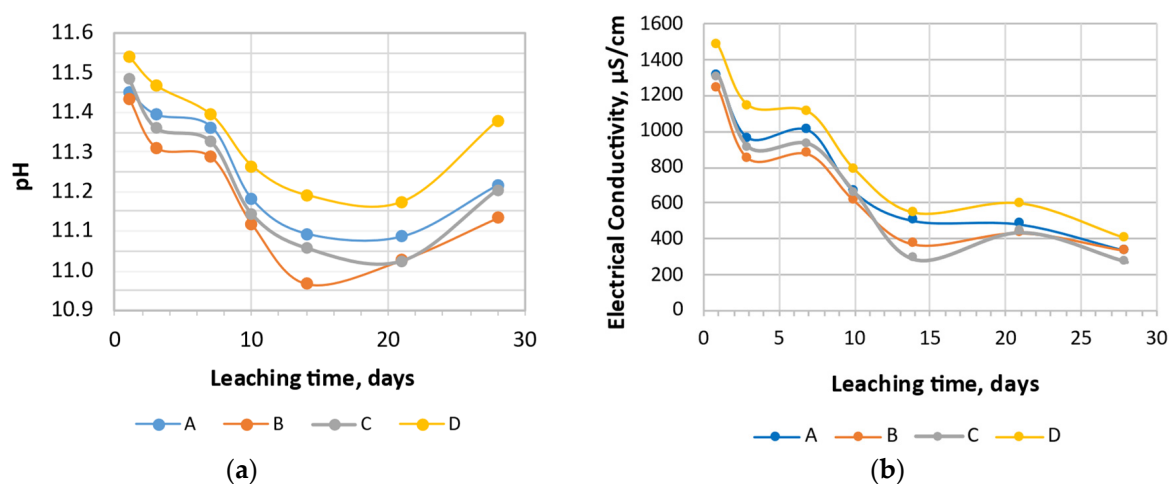


Figure 7. Changes in pH (a) and electrical conductivity (b) of the spent contact solution over time.

For composition D, the pH and electrical conductivity values are somewhat higher, which is explained by the larger content of Portland cement.

The error in the electrical conductivity measurements is ± 1.5 . The permissible limit for the pH measurement error is ± 0.01 pH units.

3.4. Ca^{2+} and SiO_2 Concentrations in the Spent Contact Solution

An illustration of the most representative results (compositions C and D) is shown in Figure 8.

An analysis of changes in the Ca^{2+} concentration shows that the intensive decalcification of the cement matrix occurs during the 3–10-day leaching period. This is followed by a gradual decrease in the Ca^{2+} concentration with a subsequent increase, possibly due to the formation of secondary compounds.

Composition D is characterized by higher Ca^{2+} concentrations throughout the experiment, which may be related to the larger proportion of Portland cement (PC/FA = 1/1.77), providing higher content of calcium-bearing hydrate phases and the formation of more soluble hydration products. Despite the greater absolute Ca^{2+} release for composition D, the relative calcium release is comparable to that of composition C.

The SiO_2 concentration decreases at the initial stage, which is associated with the retention of silicon in the structure against the background of active Ca^{2+} release. The

subsequent increase in the SiO_2 concentration in the contact solution is caused by the destruction of the C-S-H phase and the leaching of the cement silicate framework. For composition C, higher SiO_2 concentrations are observed in the 3–10-day interval, which may be associated with the higher fly ash content and, accordingly, a greater contribution of silicon-containing phases to the leaching processes.

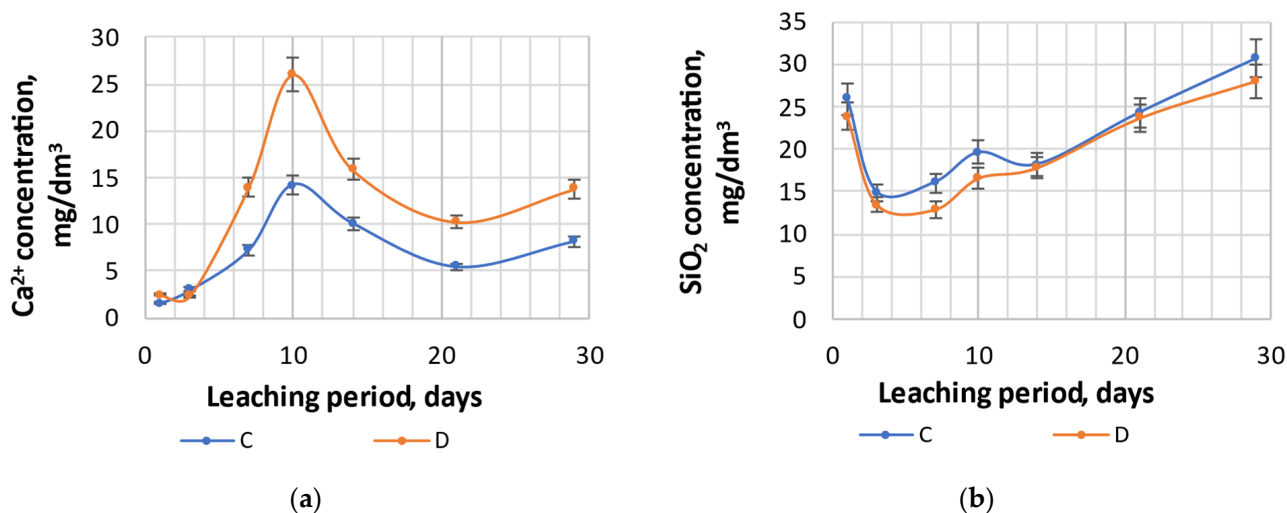


Figure 8. Changes in leaching parameters in the contact solutions over time. (a) Ca^{2+} concentration; (b) SiO_2 concentration.

From the above graphs in Figures 7 and 8, when comparing compositions C and D, it is established that increasing the fly ash content to an FA/PC ratio of 2.5 (composition C) makes the cement matrix more resistant to leaching.

4. Conclusions

This study evaluated the leaching behavior of ^{137}Cs from Portland cement–fly ash matrices containing irradiated HEU uranium–graphite fuel from the IGR reactor. Four cement matrix compositions with W/B ratios of 0.38–0.42 and FA/PC ratios of 2.5 and 1.77 were investigated.

For all cement matrix compositions, the initial ^{137}Cs leaching rates exceeded the regulatory limit of $1 \times 10^{-3} \text{ g}/(\text{cm}^2 \cdot \text{day})$ during the first 1–3 days of testing. From day 7 onward, the leaching rates decreased below this limit.

Measurements of the pH, electrical conductivity, and Ca^{2+} and SiO_2 concentrations in the leachate solutions showed that the early stage of leaching was associated with the washout of readily soluble compounds and the decalcification of the cement matrix, followed by slower changes in the silicate structure. Based on the combined ^{137}Cs leaching behavior and matrix component leachability, two candidate compositions were selected for further scale-up studies:

1. W/B = 0.40, FA/PC = 2.5 (composition B);
2. W/B = 0.42, FA/PC = 1.77 (composition D).

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Abbreviations

The following abbreviations are used in this manuscript:

FA	Fly ash
FEP	Full energy peak
HEU	Highly enriched uranium
IGR	Impulse graphite reactor
NNC RK	National Nuclear Center of Republic of Kazakhstan
PC	Portland cement
WAC	Waste acceptance criteria
W/B	Water-to-binder ratio

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