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To cite this article: Tatiana Kulagina et al 2019 IOP Conf. Ser.: Earth Environ. Sci. 227 052030

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Inclusion of liquid radioactive waste into a cement compound with an additive of multilayer carbon nanotubes

Tatiana Kulagina1, Vladimir Kulagin1,4, Eleonora Nikiforova1, Dmitriy Prikhodov1,2, Alexander Shimanskiy1 and Fengchen Li3
1 Siberian Federal University, Krasnoyarsk, Russia;
2 Radiochemical Plant of the Integrated Mining and Chemical Works, Zheleznogorsk, Russia;
3 Sino-French Institute of Nuclear Engineering and Technology Sun Yat-Sen University, Zhuhai, China.
4 Email: v.a.kulagin@mail.ru

Abstract. Long-term and safe management of radioactive wastes resulting from implementation of nuclear weapon programs still remains the key problem of nuclear energetics. The technology based on admixture of liquid radioactive wastes in inorganic hydraulic binders (cementation process) has a number of cogent advantages: the process is of low temperature, it proceeds without formation of gaseous radioactive wastes and the obtained cement compound ensures safe storage for a long time (more than $10^6$ years). A possibility of applying reinforcing fillers to increase the level of filling of the cement compound with liquid radioactive wastes was investigated. These fillers are multilayered carbon nanotubes (MCNT) and pyrogenic silica (aerosil). It has been found that the proportion of MCNT should not exceed 1.5% by weight of the cement content and the aerosil proportion should not exceed 0.5% of the binder weight. The expediency of using the effects of cavitation technology in preparation of cement compounds is shown.

1. Introduction

Russia has developed and introduced its own legal and organizational system of nuclear and radioactive safety management [1–3] in compliance with the international conventions recognized by the Russian Federation [4, 5]. The effective system of management of radioactive wastes (RAW) in Russia evolved in the process of development of nuclear technologies in the USSR, which makes it different from the systems adopted in other countries [6, 7]. The technologies of management of radioactive wastes and low-soluble sediment are reviewed in [8–12] and the extraction technology based on the PUREX process is described in [10, 11, 13–16]. The most popular approach in current use to solidifying liquid radioactive wastes of low and medium level activity is to encapsulate liquid radioactive wastes (LRW) in inorganic binders [6, 17–23].

The technology of mixed uranium-plutonium fuel produces alpha-radioactive wastes requiring special treatment. Such countries as USA, Germany, France, Japan, and Belgium conventionally use cementation to immobilize radioactive, including alpha-radioactive, wastes (RAW) [24]. This approach to conditioning liquid RAW is the most widely used technology. The process involves immobilizing LRW in a cement matrix to obtain a solid end product (cement compound), which is then delivered for further long-term and safe storage.
Cements of various types have been chosen for fixation of alpha-radioactive wastes due to the unique properties of the binding component. Cement compounds are excellent immobilizers for actinoids. Cements can be used to condition solid wastes and heterogeneous pulps. They feature high thermal stability owing to optimum technology, including low-temperature cementing. The cement compound is formed by Portland cement and thermodynamically stable hydrated salts [25]. The key requirements to the end forms of solidified RAW can be summarized as follows. They must be mechanically, physically and chemically stable enough to ensure their safe management, transportation and storage; more over the form and packaging of solidified wastes must fit the conditions of the storage place to minimize release of radionuclides into the biosphere [26].

Assuming that providing reliable natural and engineering protective barriers for solidified RAW storage will allow increasing the admissible level of individual radionuclides (primarily, actinides) in the wastes to be solidified, it is reasonable to investigate how the content of RAW in a cement compound can be increased and how this is going to affect the quality of the compound. Positive results of the research are expected to pave way for higher specific volume of buried wastes, which will cut the building and operating costs of a solid RAW storage facility.

The limiting level of individual radionuclides in a cement compound will be dictated by the nuclear safety consideration and the rate of leaching of the most soluble radionuclides (137Cs, 90Sr) because the high radiation stability of the cement matrix (cements are normally stable up to 10 MGy) suggests that the effect of radiation is not going to be the critical factor to limit the admissible level of radionuclides. A higher content of transuranium radionuclides in a cement compound will result in an insignificantly small increase in the absorbed dose (far from 10 MGy) and will not affect the quality of the cement compound. When cemented RAW are buried in metal containers, the container material will provide an additional protective barrier [27].

In terms of nuclear safety, the admissible content of radionuclides will be controlled by the content of α-emitting transuranium elements so that a possibility of development of a self-sustained nuclear fission chain reaction is ultimately excluded. To that end, a limit is imposed on the content of α-emitting transuranium elements. Their safe content in water mixtures is found as a ratio of the plutonium weight to the natural uranium weight. The nuclear-safe content of plutonium in water mixtures can be found in the tables [28, 29].

One of the parameters providing long-term and safe storage and burying of radioactive wastes is their physical-chemical property. When choosing the form of buried waste, not only its quality should meet the norms and regulations in effect but also economics considerations should be taken into account.

Several types of containers (disposable protective containers CRAD-1.36; CRAD-1.36Т and CRAD-3.0; DPC-150-1.5) and a standard 200-liter metal barrel are used to store the cement compound. Strong heating of the cement compound in the process of its hardening is possible in each of those storage options.

The nuclear safety study [29] on management of radiochemical production wastes has shown that when 200-liter containers (barrels) are used to store solidified materials, nuclear safety is ensured for any type of wastes, provided plutonium content in the package is not higher than 50 g and uranium-235 does not exceed 80 g.

From the point of view of leaching of the most soluble radionuclides 137Cs, 90Sr their reliable trapping in a cement compound is achieved by introducing sorbents into the matrix material. These sorbents are bentonite and zeolite clays (e.g. clinoptilolite) featuring high adsorption ability due to the high content (over 70 % wt.) of the clay mineral montmorillonite [30]. Clays consisting primarily of montmorillonite contain 60 to 100 mg-equiv. exchange cations per 100 g substance [31].

Condensing of the solidifying pulp by way of decantation, filtration or evaporation can improve the level of filling of the cement compound. In our present research, this was achieved by means of "dry" cementing (hardening of the solid residual of the pulp).
2. Materials
In our experiments aimed at finding the optimum composition and cementing technology, the cement compound was prepared using:

- CEM I 42.5B, GOST 31108-2003 Portland cement (former PC500-DO) by OJSS "Mordovcement";
- TU 2163-002-12763074 grinded clinoptilolite from the Kholinsk deposit as a sorbent additive of the following size composition (percentage by weight): larger than 1 mm – 2.55; up to 0.5 mm – 13.74; up to 0.25 mm – 12.20; up to 0.125 mm – 34.19; up to 0.1 mm – 15.23; up to 0.05 mm – 20.03; smaller than 0.05 mm – 2.06;
- tap water and water subjected to cavitation treatment (Table 1);
- insoluble residue of the hydroxyl pulp.

The isotopic composition and the amount of radionuclides per kg pulp are given in Table 1 and Table 2 contains data on heat release of fission products of the most gamma-active isotopes of the pulp. The total heat release from 1 kg pulp, considering all isotopes, is $1.68 \times 10^{-4}$ W [34].

| Table 1. Isotopic composition and the amount of radionuclides in 1 kg of pulp. |
|---------------------------------|-------------------|
| Uranium concentration, g/kg    | $1.0 \times 10^{-3}$ |
| Plutonium concentration, g/kg  | $2.8 \times 10^{-4}$ |
| Americium-241, Bq/kg          | $6.628 \times 10^{5}$ |
| Strontium-90, Bq/kg            | $3.7 \times 10^{7}$  |
| Cobalt-60, Bq/kg               | $6.668 \times 10^{4}$ |
| Cesium-137, Bq/kg              | $3.282 \times 10^{7}$ |
| Europium-152, Bq/kg           | $3.008 \times 10^{5}$ |

| Table 2. Heat release by fission products. |
|-------------------------------------------|
| Nuclide | Weight, g | Specific activity, Bq/g | Activity level, Bq | Specific heat release, W/g | Heat release, W |
|----------|------------|------------------------|-------------------|----------------------------|----------------|
| $^{60}$Co | 1.59E-09   | 4.19E+13               | 6.66E+04          | 1.74E+01                   | 2.77E-08       |
| $^{137}$Cs | 1.02E-05   | 3.22E+12               | 3.28E+07          | 9.58E-02                   | 9.75E-07       |
| $^{152}$Eu | 3.66E-12   | 8.19E+16               | 3.00E+05          | 1.05E+04                   | 3.83E-08       |
| Sum      | --         | --                     | 3.32E+07          | 1.04E-06                   |                |

Our studies were conducted on the pulps, their solid phase consisting predominantly of metal hydroxides and polymerized silicon acid (Table 3) as well as of manganese oxides and niobium pentoxide, ion-exchange resin residuals, nickel and cesium ferrocyanides. A considerable part of transuranium elements in the pulp is contained in the form of fine-dispersed dioxide particles [35].

Dry pulp was introduced into the compound in the amounts to fill 1.6, 4.8, and 9.6 % by weight. The water-to binder (W/B) relation was 0.7–0.9. Table 4 shows composition of the prepared cement compounds.

After trial mixture, the cement grout composition was adjusted using binding components obtained specially for the experimental purpose and the grout flow tests were conducted with a Suttard's viscometer as prescribed by GOST 23789-79 "Gypsum binders. Test methods."
Table 3. Hydroxides of stable metals in the pulp.

| Solid fraction component of the pulp | Amount in the pulp, g/l |
|-------------------------------------|-------------------------|
| Al(OH)$_3$                          | 11.27                   |
| Fe(OH)$_3$                          | 27.84                   |
| Cr(OH)$_3$                          | 2.3                     |
| Mn(OH)$_2$                          | 38.12                   |
| Ni(OH)$_2$                          | 14.17                   |
| SiO$_2$                             | 6.3                     |

Table 4. Cement compound formulations % by weight.

| Compound type index | Compound components        | Component amount |
|---------------------|---------------------------|------------------|
|                     |                           | Composition A    | Composition B    | Composition C    |
| 1                   | Portland cement           | 54.6             | 52.9             | 50.2             |
|                     | Water                     | 43.8             | 42.3             | 40.2             |
|                     | Pulp                      | 1.6              | 4.8              | 9.6              |
| 2                   | Portland cement           | 49.9             | 48.6             | 46.7             |
|                     | Bentonite                 | 5.0              | 4.9              | 4.7              |
|                     | Water                     | 43.5             | 41.7             | 39.0             |
|                     | Pulp                      | 1.6              | 4.8              | 9.6              |
| 3                   | Portland cement           | 54.5             | 53.2             | 51.0             |
|                     | S-3 Superplastificator    | 43.5             | 41.7             | 39.0             |
|                     | Water                     | 1.6              | 4.8              | 9.6              |

3. Experimental technique

We here report the results of our studies on cement compounds reinforced by carbon nanotubes and obtained by the cavitation technology [32, 33]. In order to elaborate a new method of management of radioactive wastes, a series of experiments to produce cement compounds was conducted at the laboratory facilities of the Federal State Enterprise "Integrated Mining and Chemical Plant". We studied the properties of compounds depending on the mode of preparation of the cement mixture and the pulp as well as on the properties of water that was prepared using the effects of hydrodynamic cavitation. The experimental procedure is described in [18]. Our principal tasks were:

- to see how physical chemical properties of the water influence the quality of the obtained cement compound;
- to check the effect of reinforcement by carbon multilayered nanotubes on the admissible amount of liquid radioactive wastes included in a cement compound.

The experimental compounds were prepared as follows. Clinoptilite and the hydroxide pulp in predetermined amounts were loaded into plastic cups and mixed. In 15 minutes, added were predetermined amounts of cement and water and the resultant composition was thoroughly mixed and then poured into specially prepared fluoroplastic moulds to obtain cylinder-shaped specimens, each 20 mm in diameter and height. By means of light tapping on the mould body for several minutes the mixture was compacted and the air removed. Next, the moulds with specimens were placed in a standard curing cabinet where the relative air humidity was maintained at 95 ± 5 %, the temperature being 20–30 ºC. In 24 hours the specimens were taken out of the moulds and returned into the cabinet for further 28-day curing. The cured specimens were subjected to mechanical strength tests and tests for water and frost resistance. To ensure comparability and validity of the results, characteristics of the specimens were derived from three experimental runs.
To estimate the efficacy of cavitation treatment of water, a number of physical-chemical parameters of water were monitored such as water temperature, dissolved oxygen concentration (DOC), pH index, electric conductivity, oxidation-reduction potential (ORP) and others. The measuring instruments used were a MARK-201 dissolved oxygen analyzer and a four-parameter WaterTest meter. The fundamentals of cavitation treatment of various media are described in [32, 33].

In our experiment, we used a cavitation mixer fitted with a control and power supply unit [36]. The mixer motor generated 1 kW and the useful capacity of the chamber was 3·10⁻⁴ m³. A two-bladed impeller with a wedge profile and the wedgeopening angle from 10 to 90 degrees was used as an actuator. The motor could be revved up to 14000 rpm, which allowed the cavitation number as high as χ= 0,05 to be obtained. The control circuit made it possible to smoothly control the rotor speed maintaining the number of revolutions irrespective of the changing load and to control the treatment time. In this particular case, a 20- deg. wedge profile was chosen based on the preliminary study [37]. Cavitation treatment of water was performed when the reactor was 100 % and 65 % full (Table 5).

| Filled volume of SC-reactor, V, % | Water parameters before treatment | T, °C | DOC, % saturation | S, Ohm⁻¹/cm | pH | E, mV |
|----------------------------------|----------------------------------|------|------------------|-------------|----|------|
| 100                              | 20.7                             | 40.5 | 32.6             | 85.1        | 96.65 | 4.56 |
| 100                              | 101.2                            | 34.6 | 27.2             | 87.65       | 95.7  | 5.5  |
| 65                               | 20.7                             | 34.6 | 27.2             | 87.65       | 95.7  | 5.5  |
|                                  |                                   |      |                  |             | 5.5   | 13   |
|                                  |                                   |      |                  |             | 6.0   | 5.7  |
|                                  |                                   |      |                  |             | 128.5 | 147  |

Note: DOC – dissolved oxygen concentration; α – wedge angle of the wedge-shaped cavitation of the SC-reactor; E – oxidation-reduction potential; S – electric conductivity.

In [38, 39], it was found that the high pressure (up to 1000 MPa) and intense heat (up to 15000 °C) resulting from collapsing cavitation microbubbles in water give rise to the phenomenon of mechano-thermolysis. That is, mechanical chemical reactions are initiated that produce О ₂, Н₂, Н₂О₂, ОН – during radical recombination and the decomposition results in the formation of free hydrogen bonds giving rise to chemiluminescence. The latter, in turn, affects the electric conductivity, surface tension, oxygen content, and pH index. Relaxation of the described properties takes 7–10 days, which allows these properties to be used in various technological processes right where they are or be moved any distance. Water with such properties is capable of dissolving sparingly soluble materials [32, 38]. The water characteristics are given in Table 6.

| Water sample | Parameter |
|--------------|-----------|
|              | pH        | Salinity, mg/l |
| Tap water    | 7.5       | 310            |
| Cavitation-treated water | 7.8 | 3.01 |
| Sample 1     | 7.7       | 2.64           |
| Sample 2     | 7.6       | 0.65           |

4. Results and discussion
Mechanical strength tests have shown that reducing the salt content from 3.01 to 0.65 mg/l increases the mechanical strength from 27.5 to 47.6 MPa while the pH index drops from 7.8 to 7.6 (Tables 6, 7).
Therefore, an unambiguous conclusion on the effect of the salt content on the strength properties of cement compounds is not possible. This issue requires further investigation with due consideration of all the factors involved. Based on the results of previous studies [38, 39], it is however quite reasonable to suggest that in this case the leading role is played by the improved solving ability of the water modified by cavitation water treatment, which facilitates chemical homogenization and dispersation of LRW [10–12, 38].

A chart of the mechanical strength depending on the hydroxide pulp content in cement compounds after 28 days of normal hardening is shown in Figure 1. The strength of all the samples drops to half the strength of the pure binder already after the first addition of the pulp (Composition A), Table 4.

| Index | Initial mechanical strength of cement compounds after 28 days, MPa |
|-------|---------------------------------------------------------------|
|       | Tap water | Samples with cavitationally treated water |
|       | W / C     | W / В | № 1 | № 2 | № 3 |
| 0.5   | 0.41      |       | 36.0 | 32.5 | 27.4 | 27.7 | 39.2 | 45.2 | 52.2 | 43.0 |
|       | Mean 34.2 | Mean 27.5 | Mean 42.2 | Mean 47.6 |

Note: W / C – water-cement ratio, W / В – water-binding ratio, W / В = W / (C + K-factor):

The K-factor is determined by the introduction of an additional filler in accordance with the relevant regulatory documents [2].

This happens because adding the filler (the pulp) while maintaining the same water-to-binder ratio dilutes the binder. The latter results in deteriorated surface wettability of cement particles and hence in the loss of cement strength [40]. Adding yet more pulp leads to even further deficit of the binder and eventually results in the drop of strength below the regulated value (4.9 MPa) when the filler content is increased to 9.6 % wt. (Compositions B).

The surface phenomena occurring at the phase interface affect the structure and properties of the boundary layers of the binder. Intensity of the mentioned processes is controlled by introducing various surface-active substances (SAS) that change the surface wettability of the binder and the adhesion strength at the phase interface [40]. So the increased mechanical strength of Type 3 samples is attributed to admixing a C-3 superplasticizer to their composition. The underlying mechanism is that SAS molecules interact with calcium ions on the surface of cement grains to form a surface film of the type Si-O–Ca–SO3R, which reduces the internal friction in the concrete mixture. Moreover, due to its peptization ability the C-3 admixture prevents formation of flocculi from cement particles in the process of hydration thereby increasing the specific surface area of the particles and facilitating the processes of hydration and structural formation of a cement stone.

Adding bentonite (Type 2) leads to additional dilution of Portland cement and hence to yet smaller portion of the binder in the compound, which accounts for the reduced mechanical strength compared to the pure compound (Type 1).

Frost resistance was assessed by comparing the mechanical strength of the specimens after 30 freeze-thaw cycles against their initial strength. Frost resistance is considered acceptable if the strength of specimens after testing is not less than 75 % of their initial strength (but no less than the admissible 5 MPa). The frost resistance tests were conducted in an Espec SH-242 temperature and humidity chamber and the results are summarized in Table 8.

The rate of leaching was measured following the procedure prescribed by GOST 52126-2003. The prepared specimens were removed from the standard curing cabinet, placed into plastic cups and poured over with 50 ml distilled water. The relation of the contact solution volume to the specimen surface was 2.65. The cups were covered to stop water evaporation and left to sit in a fume cupboard.

The contact solution was removed and sent for testing while the specimens were flooded with a new portion of distilled water. The contact solution was tested for Cs-137 concentration applying the
techniques conventionally used at the Federal State Unitary Enterprise "Integrated Mining and Chemical Works". Tests were repeated until a constant value of the rate of leaching had been obtained. The latter was found as

$$R_n = \frac{a_n}{A_0 \cdot S \cdot \tau_n}$$  \hspace{1cm} (1)

where $R_n$ is the rate of leaching of cesium-137, g/(cm$^2$-day.); $a_n$ is the activity of cesium-137 that has leaked into the water over the time $\tau_n$, Bq; $A_0$ is the specific activity of cesium-137 in the specimen, Bq/g; $S$ is the open geometric surface area of the specimen contacting the water, cm$^2$; $\tau_n$ is the length of the $n$-th period of leaching, days. The measured rates of leaching for cesium-137 (obtained with the use of cavitation technology) are given in Table 9 and they do not exceed the documentarily regulated values in the atomic industry.

![Figure 1. Mechanical strength of cement compound specimens versus pulp content.](image)

**Table 8.** Frost resistance test results for the cement compound specimens.

| No  | Initial compressive strength of the compound, MPa | Mean, MPa | Compound strength after frost resistance tests, MPa | Mean, MPa | Changes in the mechanical strength, % |
|-----|-------------------------------------------------|-----------|-------------------------------------------------|-----------|--------------------------------------|
| 0-1 | 36.0                                            | 34.2      | 32.8                                            | 29.8      | -12.9                                |
| 0-2 | 32.5                                            | 27.4      | 27.7                                            | 22.0      | -21.5                                |
| 1-1 | 27.4                                            | 39.2      | 32.8                                            | 39.2      | 0.5                                  |
| 1-2 | 27.7                                            | 45.2      | 42.2                                            | 42.4      | 0.5                                  |
| 2-1 | 2-2                                             | 3-1       | 3-2                                             |           |                                       |
| 3-1 | 52.2                                            | 47.6      | 44.0                                            | 48.4      | 1.7                                  |
| 3-2 | 43.0                                            | 45.2      | 34.1                                            | 42.4      | 0.5                                  |
Table 9. The leaching rate of Cs-137.

| Specimen No | $A_0$, Bq/g | $R_{n}$, g/(cm²·day) |
|-------------|-------------|-----------------------|
|             | Number of days | 1 | 3 | 7 | 10 | 14 | 21 | 28 | 56 |
| 0-1         | 5850 \quad 1.03\cdot10^{-2} \quad 2.47\cdot10^{-3} \quad 2.93\cdot10^{-4} \quad 2.00\cdot10^{-4} \quad 1.88\cdot10^{-4} \quad 1.55\cdot10^{-4} \quad 9.46\cdot10^{-5} \quad 4.89\cdot10^{-5} |
| 0-2         | 980 \quad 9.4\cdot10^{-2} \quad 2.78\cdot10^{-2} \quad 3.04\cdot10^{-4} \quad 2.10\cdot10^{-4} \quad 1.55\cdot10^{-4} \quad 1.33\cdot10^{-4} \quad 9.01\cdot10^{-5} \quad 5.67\cdot10^{-5} |
| 1-1         | 1840 \quad 6.12\cdot10^{-2} \quad 1.51\cdot10^{-2} \quad 2.01\cdot10^{-3} \quad 1.13\cdot10^{-3} \quad 8.47\cdot10^{-4} \quad 6.18\cdot10^{-4} \quad 5.09\cdot10^{-4} \quad 2.34\cdot10^{-4} |
| 1-2         | 3840 \quad 6.69\cdot10^{-2} \quad 3.29\cdot10^{-2} \quad 1.87\cdot10^{-3} \quad 1.16\cdot10^{-3} \quad 8.47\cdot10^{-4} \quad 5.83\cdot10^{-4} \quad 5.09\cdot10^{-4} \quad 2.34\cdot10^{-4} |
| 2-1         | 2050 \quad 5.07\cdot10^{-2} \quad 2.82\cdot10^{-2} \quad 1.83\cdot10^{-3} \quad 1.45\cdot10^{-3} \quad 7.00\cdot10^{-4} \quad 6.18\cdot10^{-4} \quad 5.23\cdot10^{-4} \quad 2.55\cdot10^{-4} |
| 2-2         | 3150 \quad 7.95\cdot10^{-2} \quad 4.36\cdot10^{-2} \quad 2.16\cdot10^{-3} \quad 1.67\cdot10^{-3} \quad 7.31\cdot10^{-4} \quad 6.62\cdot10^{-4} \quad 5.18\cdot10^{-4} \quad 2.71\cdot10^{-4} |
| 3-1         | 3680 \quad 7.34\cdot10^{-2} \quad 3.78\cdot10^{-2} \quad 2.86\cdot10^{-3} \quad 1.91\cdot10^{-3} \quad 1.81\cdot10^{-3} \quad 1.81\cdot10^{-3} \quad 1.81\cdot10^{-3} \quad 1.81\cdot10^{-3} |
| 3-2         | 1400 \quad 1.22\cdot10^{-1} \quad 6.07\cdot10^{-2} \quad 3.45\cdot10^{-3} \quad 2.13\cdot10^{-3} \quad 1.74\cdot10^{-3} \quad 8.41\cdot10^{-4} \quad 7.19\cdot10^{-4} \quad 3.57\cdot10^{-4} |

Of all the variety of highly efficient chemical and mineral admixtures, the nano-sized admixtures have lately been of a particular practical interest [41–45]. The developed specific surface area associated with the small particle size makes them chemically active and allows them to be classified as super-pozzolana. The amount of carbon fulleroid-type nano-admixtures (single- and multilayered carbon nanotubes) required to modify the concrete properties is about 5.0 % of the cement weight [42]. Carbon nanotubes are cylinder-shaped molecules consisting of a folded graphite plane with the diameter ranging from 0.8 to 5.0 nm (Figure 2) [43]. The amount of nanotubes was up to 5.5 % wt. of the cement content. From Figure 3 it follows that the mechanical strength grows by 37 % upon adding 1.5 % wt. of carbon nanotubes.

![Figure 2. Electron micrograph of a multilayered carbon nanotube.](image1)

![Figure 3. Mechanical strength of cement compound samples depending on the number of multilayered carbon nanotubes added.](image2)
Figure 4. Distribution of mechanical stresses in the space around a carbon nanotube.

Improvement of the mechanical properties of a material due to carbon nanotubes is associated with the two effects. First, intense interaction of nano-sized filler particles with the cement matrix material gives rise to a considerably larger number of crystallization centers (nuclei), which eventually leads to formation of a material with a considerably higher degree of crystallinity or to formation of a material with a fine crystalline structure, enhancing thereby the mechanical strength of the material [44]. Second, the elongated shape of nanotubes generates asymmetric strain (Figure 4) in the hardening material, which results in the formation of oriented clusters, cybotaxic groups and crystalline arrangements of macromolecules [45].

The observed in Figure 3 loss of strength upon adding carbon nanotubes in excess of 1.5 % wt. is associated with the low amount of the binder which is insufficient for the highly developed surface area of carbon nanotubes. The high surface energy of multilayered carbon nanotubes promotes formation of globules. These globules are difficult to distribute uniformly in a water medium, which calls for special dispergation technologies (such as ultrasound exposure, cavitation and others).

5. Conclusions
The mechanical strength of a cement compound has been found to be independent of the contained amount of individual radionuclides. It is primarily dependent on the amount of radiochemical production wastes (pulp). At elevated levels of RAW in a compound its strength will be controlled by the portion size of the binding material. Application of various modifying admixtures further contributes to the decrease in the portion of the binder in the compound and reduces its strength.

The suggested application of carbon nano-sized admixtures of a fulleroid type (multilayered carbon nanotubes) has improved the compound strength by 37%. This has been achieved by adding just 0.5 % carbon nanotubes of the cement weight and the use of mechanical dispergation. Yet, further research is required to study the influence of MCNT added to a compound with an increased pulp content on the rate of leaching, frost resistance and long-term exposure to water.

A noticeable influence of cavitation-activated water on the characteristics of a cement compound in the process of its preparation has been proved by the obtained findings on immobilization of radioactive wastes in a cement matrix [21–23]. Employing cavitation effects for compounding radiochemical production wastes increases the strength of the cement stone 2–3 times against the conventionally prepared solution and improves its resistance to thermal cycles (freeze-thaw resistance) without affecting the rates of leaching of radionuclides from the cement matrix, which remain within the regulated range of values adopted in the nuclear industry. The latter characteristic is one of the key factors determining reliability and safety of long-term storage and burying of radioactive wastes.

The radius of the pulp particle decreases with increasing number of ultrasonic treatment cycles from initial 50 µm to 10 µm after six cycles. Application of the suggested methods of management of radioactive wastes allows their utilization efficiency to be improved twice as much due to the synergetic effect of dispergation of low soluble sediments and cement stone and formation of a stronger cement compound structure. In the process of treatment of the “cement milk” a needle-like structure of the cement stone is formed that alters the behavior of the crystal-chemical reaction of
concrete hardening and acts as centers of directional crystallization. This, on the one hand, results in a fibrillary structure of the cement stone and, on the other hand, gives rise to a reinforcing oriented supramolecular structure.

Acknowledgment
The reported study was funded by RFBR and the government of Krasnoyarsk region according to the research projects №№ 17-48-240386, 18-48-242001 and 18-41-242004.

References
[1] On the Use of Nuclear Energy: RF Federal Law of 21.11.95 No 170-FZ (Ed. of 02.07.2013 with amendments effective of 02.09.2013).
[2] Safety Management in Russia. Legal, Socio-Economic and Scientific and Technological Aspects 2003 (Moscow: Znanije) 400
[3] Sanitary Guidelines for Radioactive Waste Management (SPORO-2002). SP 2.6.6.1168-02. 2003 (St. Petersburg: Dean) 64
[4] Joint convention on safety management of spent fuel and safety management of radioactive wastes. Vienna, September 5, 1997. Effective date for Russia 19.04.2006. http://www.bellona.ru/Casefiles/vienna97
[5] Convention on Nuclear Safety. Vienna, June 17, 1994. Effective date for Russia 24.10.1996. http://www.bellona.ru/Casefiles-/vienna94
[6] Klyuchnikov A A, Pazukhin E M, Shigera Yu M, Shigera V Yu 2005 Radioactive wastes of nuclear power plants and their management technologies ed Yu M Shigera. (Kiev: Chernobyl) p 496
[7] Lebedev V M Nuclear Fuel Cycle 2005 (Moscow: Energoatomizdat) 305
[8] Volkov V G and Chesnokov A S 2011 Industrial Journal. No 11–12
[9] Milyutin V V, Guelis V M 2011 Modern approaches to treatment of liquid radioactive wastes and radioactive-contaminated natural waters (Moscow: IFHE RAS)
[10] Kulagina T A, Kulagin V A, Matyushenko A I 2014 Technosphere safety in atomic power engineering (Krasnoyarsk: Grotesk Publ., SFU) 286
[11] Kulagina T A, Kozin O A, Matyushenko A I 2015 Environmental safety of technosphere projects (Krasnoyarsk: Grotesk Publ) 323
[12] Kulagina T A, Kulagin V A, Matyushenko A I 2013 J. Sib. Fed. Univ. Eng. technol 6(2) 123–149
[13] Todd T 2009 Introduction to Nuclear Fuel Cycle Separations: CRESPP Short Course – Introduction to Nuclear Fuel Cycle Chemistry (Crystal City, VA)
[14] http://www.osti.gov/scitech/biblio/5220059
[15] Tanayev I G, Myasoyedov B F 2002 Chemical behavior of plutonium in alkaline wastes: forms of existence, redox reactions and structural transformations. (Moscow: V I Vernadsky Institute of Geochemistry and Analytical Chemistry RAS) 356
[16] Kasatkin A G 1973 Basic processes and equipment of the chemical technology (Moscow: Chemistry) 750
[17] Kulagina T A, Popkov V A 2015 J. Sib. Fed. Univ. Eng. technol 8(2) 198–207
[18] Kulagina T A, Popkov V A 2015 J. Sib. Fed. Univ. Eng. technol 8(3) 362–368
[19] Yermolayev V M, Zakharova E V, Mironenko M V 2006 Radiochemistry 47(3) 374–379
[20] Shilov V P, Fedoseyev A M 2003 Radiochemistry 45(5) 441–444
[21] Kulagina T A, Kulagin V A, Moskvichev V V, Popkov V A 2016 Ecological Environment and Industry of Russia 20(10) pp 4–10 DOI: 10.18412/1816-0395-2016-10-4-10
[22] Kulagina T A, Kulagin V A, Popkov V A 2016 J. Sib. Fed. Univ. Eng. technol. 9(2), pp 280–295 DOI: 10.17516/1999-494X-2016-9-2-280-295
[23] Kulagina T A, Popkov V A 2015 J. Sib. Fed. Univ. Eng. technol. 8(7) 917–927 DOI: 10.17516/1999-494X-2015-8-7-917-927
[24] Aloi A S 2016 Analytical report on the methods of management of alpha-containing RAW (St. Petersburg) 34
[25] Technological and Organizational Issues of Radioactive Wastes Management 2005 (Vienna IAEA-TCS-27, ISSN 1018–5518) 230
[26] NP-019-2015 Collection, processing, storage and conditioning of liquid radioactive wastes. Safety Requirements 2015 (Moscow) p 13
[27] Pugliese et al 2010 Nuclear Eng. Design
[28] PBYa-06-02-93 Nuclear industry safety rules for "B" and "DB" type plants 1993 (Moscow) p 36
[29] Vnukov V S, Sichkaruk O V, Chkuaseli L I 2004 Atomic Strategy 13 1–6
[30] Yezhkov V O, Yezhkova A M, Yapparov A Kh 2013 The technology of using bentonite clays as sorbents of heavy metal salts from animals in the regions of technogenic burden in Kazakhstan Republic (BIBKOM) 100
[31] Betekhtin A G 1951 Mineral Science Course (Moscow: Geol. Lit. State Publ.) 543
[32] Ivchenko V M, Kulagin V A, Nemchin A F 1990 Cavitation Technology ed Acad. G V Logvinovich (Krasnoyarsk: KSU) 200
[33] Demidenko N D, Kulagin V A, Shokin Yu I, Li F-Ch 2015 Heat-and-mass transfer and supercavitation (Novosibirsk: Nauka) 436
[34] Database Nuclides 2000 An Chart of Nuclides, Version 1.00, European Communities, 1999.
[35] Yermolayev V M, Zakharova E V, Mironenko M V et al 2006 Radiochemistry 47(3) 374–379
[36] Kulagin V A High-Speed Hydrodynamics 1992 (Krasnoyarsk: KrPI) 134–140
[37] Kulagin V A, Sapozhnikova E S, Stebeleva O P 2014 J. Sib. Fed. Univ. Eng. technol 7(5) 605–614
[38] Kulagin V A 2004 Methods and means of technological processing of multicomponent ores with the use of cavitation effects 2004 (Krasnoyarsk: Diss. Dr. Tech. Sci.) 406
[39] Margulis M A 1986 Sonochemical reactions and sonoluminescence (Moscow: Chemistry) 288
[40] Korolev E V, Samoshin A P, Smirnov V A et al 2009 The techniques and algorithm to synthesize radiation-protective materials of new generation (Penza: PGUAS) 130
[41] Cwirzen A et al 2009 Advances in Cement Research 21 141–146
[42] Khizin V G, Starovoitova I A and Maisuradze N V 2013 Building materials 2 4–10
[43] Pukharenko Yu V, Aubakirova I U, Nikitin V A, Staroverov V D 2008 Intern. Congress Proc. “Science and innovations in building engineering” “SIB-2008” Challenges in modern construction materials science and technology (Voronezh) 1(2) 424–429
[44] Poole Ch and Owens F 2006 Nanotechnologies (Moscow: Technosphere) 336
[45] Castrillon M, Garcia C, Paucar C 2007 Dyna Medellin Colombia 152 159–165