 doi:10.20944/preprints202205.0198.v1

Article

Recycled Radioactive Waste in the Space Industry

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Abstract: The article discusses an alternative way of recycling radioactive waste (RW), presented in the form of radioactive building materials - concrete and reinforced concrete structures and metal fittings, with the further use of materials, obtained during recycling, in the space industry. That is, it is supposed to send radioactive waste into space not as a passive ballast, but as a payload that will operate in space under conditions of increased radiation.

Keywords: radioactive waste; radioactive waste recycling; industrial slag; industrial ash; aerospace industry

1. Introduction

Recycling and storage of radioactive waste is an expensive technology, in which today there are many unsolved problems. The cost of RW recycling and storage is constantly growing. Thus, in [1] the costs for the construction of 1 m^3 of RW storage are 300$ (data for 2012). RW recycling technologies are being improved, but radioactive material remains largely unclaimed. At the same time, there are industries where there is a high level of natural radiation, such as space industry. To date, nowhere has anyone considered the possibility of using radioactive waste as materials for the manufacture of electronic elements, parts and devices of space probes and planetary rovers, because these installations are designed to operate in conditions of increased radiation, which means that their own even very high radioactivity will not be a problem in their functioning.

The article discusses the current state of affairs with RW recycling and storage and assesses the possibility of RW recycling for further use of materials obtained from RW recycling in the space industry.

2. Overview of technologies for the recycling of radioactive waste, industrial slag and ash

In [2] – [5] the continuous improvement of the methods of utilization and neutralization of thorium and radium containing wastes obtained in the mining industry and during the recycling of radioactive waste is pointed out. Thus, 2% (by mass) of thorium and radioactive elements of the thorium series (mainly radium) accumulates in waste from the extraction of rare earth elements from loparite ores. Technologies have been developed to increase the concentration of thorium and radium from solutions with bringing the precipitate to a solid, non-dusting, water-insoluble, radiation-safe state, convenient for storage without sending it to a special waste storage (SWS) [3]. Liquid waste releases are decontaminated, release radionuclides and convert them into solid blocks, chemically inactive and mechanically resistant, radiation-safe, convenient for storage without being sent to SWS [4]. A facility has been developed that makes it possible to obtain a precipitate with thorium series radionuclides from solutions from the leaching of radioactive waste, and the solution is completely decontaminated and goes with wastewater into the industrial sewer [5]. The precipitate is sent for storage in the SWS. The economic effect of savings on waste transportation and construction of SWS is 100 000 $/year (data for 2002).

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In [6], it is indicated that oil sludge (waste) contains a significant amount of radionuclides. Sludge collectors and oil sludge pits accumulate 5000 m^3/year of waste and sediments contaminated with radionuclides. These sludges contain heavy metals – up to 1% by weight of the inorganic mass of the sludge. Mining and recycling of ore raw materials lead to the accumulation of uranium and thorium in wastewater – up to 1 mg/l and 0.8 mg/l, respectively. The content of yttrium and lanthanides correlates with uranium and thorium in wastewater – up to 5 mg/l and 7 mg/l, respectively.

Article [7] describes a technology for recycling liquid radioactive waste from radiochemical plants with a large amount of complexing agents of low activity (~10^5 Bq/l) using phytosorbents (obtained from malt sprouts and sawdust) that accumulate all radiation sources, except for neutron sources. Phytosorbents are burned, the volume of waste is reduced by a factor of 300, the ash is vitrified or cemented and sent to burial place. The authors of [8] describe a method for obtaining a cellulose-inorganic sorbent based on iron ferrocyanide as the best cesium radionuclide sorbent.

In [9], the composition of natural radionuclides in ash and slag from coal combustion is considered. There is an increase in the specific activity of combustion products: slag and ash. The highest activity is observed in aerosol emissions and fly ash, which have the smallest particle size.

In [10], the distribution of radionuclides between the phases of glassy materials from the recycling of ash from the combustion of spent sorbents, soil based on clay and sand was considered. The share of crystalline and metallic phases in them is 10 – 20 wt. %, the rest is glass. Cesium and strontium are predominantly in the glass phase.

To date, methods have been developed for the industrial recycling of ash in order to extract useful elements and compounds from it. In [11], the recycling of ash containing the main component - carbon and impurities of lead, boron and manganese is considered. Oxidation in a stream of oxygen and steam produces the main process gases: CO, CO_2, H_2, as well as oxides of harmful substances, which have lower toxicity limits: PbO_2, BO_2, MnO. The parameters of the reactor for this recycling have been calculated [11]. In [12], the methods of magnetic separation of iron oxides (magnetite $FeO{\cdot}Fe_2O_3$) from ash and slag from coal combustion at a condensing power plant are considered. The separation was carried out on an electromagnetic roller separator, with the aim of using magnetite as an additive in the charge for blast-furnace iron smelting at a metallurgical plant. The content of magnetite in coal ash is 6.43%.

In [13], the release of vanadium from ash from the combustion of fuel oil is considered. The analysis of ash for the composition of minerals is carried out by the X-ray phase method, the isolation of vanadium is carried out by leaching with water and sulfuric acid, followed by precipitation of 95% of vanadium.

In [14], the possibility of extracting rare earth elements from ash from the incineration of municipal solid waste was considered. It is noted that the composition of this ash for the content of rare earth elements meets the recommended standards for their industrial extraction from technogenic raw materials and from natural sources. The possibility of using the residue from recycled ash depleted in rare earth elements as a filler for composite building materials is indicated, since the content of the residue is similar to the composition of the charge used for the same purposes.

In [15], the composition of ash and slag dumps from the combustion of fuel from thermal power plants is considered. The main component is silica 30 – 60%, aluminum oxide 15 – 28% and iron oxide 3 – 7%. The rest is oxides of magnesium, calcium, potassium, titanium, aluminosilicate. The possibility of further recycling for the purpose of industrial extraction of useful elements is noted.

From the above review, we see that modern technologies make it possible to process ash from the combustion of various products with the extraction of useful elements from the ash for their use in industry. These technologies can be applied to the ash obtained from the combustion of radionuclide sorbents in order to isolate the necessary elements

 doi:10.20944/preprints202205.0198.v1

for their use in industry, including those industries where the radiation background is high.

Article [16] indicates the presence of several million tons of radioactive waste. 10000 tons of metallic radioactive waste is released from one gigawatt PWR. The cost of storage of solid radioactive waste is growing – 1000$/m^3. The cost of pure non-ferrous and highly alloyed metal is 1000$/ton (data for 2008). The cost of recycling by melting is 250$/ton. It is indicated that there are a large number of factors that determine the distribution of radionuclides between the products of metal recycling by smelting - melt, slag, gaseous fraction. In particular, ^{60}Co remains in the melt by 95%, ^{134,137}Cs is distributed equally between the slag and the gas fraction and practically disappears in the melt, ^{90}Sr passes into slag by 95%, into gas by 2%, and 3% remains in the melt. On average, slag and gas make up 10% and 1% of the mass of the original metal charge, respectively.

In [17], the technology of plasma recycling (using a plasma torch) in shaft furnaces of solid (combustible and pressurized) and liquid low- and medium-level radioactive waste is considered. The productivity of such modern installations for the recycling of solid RW is 1040 kg/h, liquid RW is 600 kg/h. The output product is vitrified slag from non-combustible waste parts and synthesis gas (H_2, CO, CO_2). The radioactive component of the synthesis gas can be converted to slag by 90% by filtering and evaporating the salt precipitate. From 56% to 85% of radioactive cesium and from 96% to almost 100% of radioactive cobalt pass into the slag from the initial waste. The cost of recycling is about 250$ per cubic meter of waste. For British conditions, the plasma method for recycling wet radioactive waste of medium activity with a volume of 30000 m^3 costs 1.3 billion $, taking into account all costs, including the entire storage life cycle. This is 4.6 times cheaper than conventional combustion and cementing.

In [18], an analysis is made of existing installations for the induction melting of RW in a glass melt for the purpose of vitrification of RW with subsequent disposal. It is noted that there are installations (in Marcoule and Grenoble, France) that allow the production of glass in the amount of 200 kg/h from solid RW and 50 kg/h from liquid RW. At the same time, in [19], the melting and casting of radioactive metals in a vacuum resistance furnace is considered, which provides a more uniform temperature field in the area of melting and heating in comparison with vacuum induction furnaces.

Reference [20] considers the recycling of metal radioactive waste (MRW). It is noted that radioactive contamination can penetrate deep into the metal and is not completely removed during etching and machining. It is proposed to use a two-stage recycling of MRW. The first stage is remelting with deep mixing in various types of furnaces. The radioactive component is concentrated in the slag, which is removed for storage. The second stage – a consumable electrode for electroslag remelting (ESR) is made from partially deactivated metal. With ESR, the electrode is melted in a flux (slag) of a certain brand, nonmetallic impurities and a radioactive component are released from the molten metal of the electrode. The metal itself flows into the mold under a layer of slag and crystallizes. The remaining radioactive component in the metal is evenly distributed over the volume of the metal blank. The deactivated metal thus obtained can be used industrially without restriction. For 300 kg of MRW, 12 kg of slag is obtained from two stages of remelting, including spent flux at the ESR stage.

In [21], the process of decontamination of low-alloy steels by melting in plasma-arc furnaces of a new generation (PAF-NG) is considered, which melt 12 tons in 45 – 60 minutes. Prior to entering the PAF-NG melter, radioactive scrap undergoes surface decontamination in an annealing furnace at 850 – 900°C, followed by descaling at a vibroabrasive unit. Using the developed technologies, in this way it is possible to reduce the surface activity of scrap by 100 times. Further remelting in PAF reduces contamination at high temperatures of the melt, since the radioactive component passes into the slag and the radioactive residue is uniformly distributed over the volume of the melt.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 16 May 2022 doi:10.20944/preprints202205.0198.v1

In [22], mathematical modeling of the process of radioactive waste incineration in a shaft furnace using a plasma torch is carried out. These furnaces provide complete combustion of radioactive waste to ash from the non-combustible part of the waste and to the gaseous state from the pyrolysis of cellulose and lignin (the combustible component of the waste). The gaseous part of the waste with radioactive cesium is captured by sorbents in the upper part of the furnace.

In [23], the formation of radioactive aerosols during welding of surface contaminated metals is considered. It is noted that when samples of such metals are calcined in a muffle furnace, they are completely deactivated during several cycles of calcination and additional mechanical removal of the surface layer. Radioactive isotopes ^{137}Cs and ^{90}Sr pass into a dispersed aerosol with a particle size of 0.1 – 20 μm. The specific activity of aerosol particles A_s after each calcination cycle depends on the calcination time of the samples and the number of calcination cycles, the calcination temperature, and is determined by the formula:

A_s [Bq/mg] = $0.2 \cdot (A_0[\text{Bq/cm}^2])^{0.59}$,

where A_0 – initial surface activity of the sample before the next calcination cycle.

Technologies for recycling slags in order to isolate useful elements or compounds for their use in industry are well developed. In [24], the technology for recycling slag from aluminum smelting and remelting is described. Slag recycling included its drying, grinding in a ball mill, roasting and finishing grinding to obtain micropowders of two types - with a particle size of up to 50 microns and more than 50 microns. The latter was used to apply wear-resistant coatings for machine parts. The composition of the slag is more than 50% corundum α–Al_2O_3 and up to 20% silica SiO_2, the remainder is oxides of iron, magnesium, calcium and titanium, which have high hardness.

In [25], a method for separating tungsten from slag from the smelting of tin raw materials is described. The slag is presented in the form of granules 0.5-4 mm in size, which is treated with a solution of soda, sodium hydroxide, sodium chloride, heated to 90°C, in the presence of vibrations of 50 Hz with an amplitude of 4 mm and spark discharges with a voltage of 30 kV with oxygen supply. Tungsten is leached into a soda-alkaline solution in the form of tungstic acid. 96% of all tungsten bound in the slag goes into solution after 3 hours of slag treatment (in the article – 50 g of slag with a grain size of 1 – 2 mm, containing 1.8% W, 23.4% Fe, 43.4% SiO_2, 13.7% CaO). The tungstic acid is then recovered from the solution by known methods.

In [26], the technology for producing titanium dioxide from ilmenite smelting slags is described. Samples of titanium slag were taken weighing 10 g, the content of the sample was 82.23% TiO2, 4.97% Fe, 2.66% SiO_2, 0.62% Cr_2O_3, 5.89% Al_2O_4, 0.63% Mn, 0.35% CaO, 0.3% MgO, 0.071% S and 0.022% P. The slag was crushed to a particle size of less than 50 μm, mixed with soda, sintered at 900°C, leached in water at 80°C, the precipitate was treated with a hydrochloric acid solution at 103°C, the TiO_2 precipitate was dried and calcined at 900°C. More than 97% of titanium dioxide was isolated from slag by this method, more than 67% of silica.

In [27], the processing of slag from the production of non-ferrous metals, which is a multicomponent system of ferrous 40%, non-ferrous metals 23%, oxides of alkaline earth metals 1% and silicon 36%, is considered. The emphasis is on the complete extraction of non-ferrous metals - zinc and copper. Leaching of non-ferrous metals is carried out first with sulfuric acid, and then with an ammonia-ammonium solution. Almost complete extraction of zinc and copper is observed.

In [28], the technology for obtaining iron silicides from slags is considered. An important role in microelectronics is played by modified iron silicides, which exhibit ferromagnetic (α-Fe_3Si and ε-Fe_5Si_3), semiconductor (β-$FeSi_2$), and phosphor (ε-Fe_2Si_5) properties. Slag for the production of ferrosilicides - copper waste (Fe_2O_3 – 63.7%, SiO_2 – 27.5%, CuO – 5.6%, Al_2O_3 – 1.2%, BaO – 0.6%, ZnO – 0.6%, PbO – 0.3%, K_2O – 0.2%, which corresponds to 44.59% Fe and 12.83% Si in converter copper slags) and molybdenum slag (SiO_2 – 78.9%, Fe_2O_3 – 10.5%, CaO – 4.2%, Al_2O_3 – 4.1%, K_2O – 0.8%, BaO – 0.7%, TiO – 0.2%,

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CuO – 0.2%, ZnO – 0.1%, which corresponds to 7.35% Fe and 36.82% Si in molybdenum slags). The slag was taken crushed to particles up to 2 mm in size. Both slags were mixed - 1 part of copper slag was taken for 3 parts of molybdenum slag, and a charge was obtained. The mixture was placed in quartz sand inside the electric furnace, heated to 2500°C for 15 s – aluminothermic reduction. The metal phase of the silicide was collected at the bottom of the slag – 65.2% - Fe, 34.78 % - Si, 0.02 % - Al. The total yield of the metal phase from the charge is 94.02%.

From the above review, it can be seen that the existing technologies make it possible to almost completely deactivate the metal during remelting, radionuclides pass into slag and gas fractions [20], [21]. Existing technologies make it possible to isolate the desired component from the slag or decompose the entire slag into components for their use in industry. This technology can be transferred to the recycling of radioactive slag, vitrified and/or cemented RW. The gas fraction can be deactivated with the help of sorbents, the combustion of which to ash will make it possible to apply the already mentioned recycling technologies to the ash.

Building concrete, reinforced concrete and metal structures contaminated with radioactive emissions accumulate as a result of the dismantling of structures of nuclear facilities, nuclear accidents, nuclear weapons testing, man-made disasters. According to the level of specific activity, these wastes are classified as low-, medium- and high-level radioactive waste. Metal structures can be assumed presented by ordinary cast iron and steel with an iron content from 95%. Concrete, as an artificial rock, contains silicon, oxygen and aluminum as the main components, the content of which is approximately the same as in natural rock: bound oxygen up to 50%, silicon up to 30%, aluminum up to 10% [29]. In order to talk further about the methods of recycling and using this radioactive building material in industry, let us consider where the above-mentioned main components of building radioactive waste - silicon, oxygen and iron - are used in industry.

3. The use of silicon Si and silica SiO_2 under high radiation conditions

It is known that silicon Si and silicon dioxide SiO_2 are the main components of solar cells, components of radio- and microelectronics, and optics [30], [31]. These devices are widely used on spacecraft and planetary rovers (martian and lunar rovers) exposed to cosmic radiation in outer space or on the surface of the Moon or Mars. [32]. Another application of silicon and silicon dioxide in conditions of increased radiation is nuclear medicine. Silicon and silicon dioxide nanoparticles with a diameter of 1 to 100 nm are used for cancer radiation therapy and tumor imaging [33], [34]. For these purposes, a radioactive label is attached to the nanoparticle by biochemical methods – one of the radioisotopes of iodine $^{131,125,124}I$ [35] or a radioisotope of rhenium ^{188}Re [33]. The next application of silicon and silicon dioxide is the use of their nanoparticles as fillers of nanocomposite materials based, for example, on polymers. It has been established that the physical characteristics of nanocomposites after exposure (and during exposure) to radiation (by gamma quanta, electrons, and ion beams) are significantly improved (up to 70% [36]) compared to non-irradiated ones. Silica micropowders are used in lithium-ion batteries [37]. To date, no attempts have been made to make nano- or micro powders initially radioactive, that is, from radioactive materials, in particular, from radioactive waste.

4. The use of iron in conditions of high radiation

The main industries in which iron-based alloys are used in conditions of high radiation are the space industry, fusion and fission reactors, oil, gas and mining industries. Iron nanoparticles and its oxides are used in electronics due to their magnetic properties for various types of sensors and as fillers in composites [38]. In nuclear medicine, iron oxide nanoparticles are used for radiation therapy and tumor imaging [39]. To create a radioactive label, iron oxide nanoparticles are irradiated with protons with an energy of 20 MeV

 doi:10.20944/preprints202205.0198.v1

at accelerators, which leads to the formation of ^{99m}Tc, ^{55,56,57,58}Co radionuclides on the surface of iron oxide nanoparticles. Powders of iron and its oxides of various fineness are widely used in electronics and robotics [40], in additive and powder technologies for the manufacture of structural parts of machines and mechanisms [41], [42], [43], [44], permanent magnets [45]. Attempts to obtain powders of iron and its oxides initially radioactive from radioactive materials and radioactive waste have not been undertaken.

It should also be noted that it is planned to create nanoporous steels that are insensitive to radiation exposure [46]. Nanoporous steels are planned to be used under conditions of increased radiation in thermonuclear and fission reactors, since irradiation will not lead to deterioration of their mechanical properties [46], which means that they can be made from radioactive waste and used also on space probes and planetary rovers.

5. Extraction of oxygen and silicon from (reinforced) concrete.

Recycling conventional concrete and reinforced concrete for industrial reuse is well established [47], [48], [49]. As well as for ordinary concrete, when recycling radioactive concrete and reinforced concrete, for crushing (and extracting reinforcement), you can use a mechanical concrete breaker with a hydraulic hammer, which must be automated or remote-controlled, work indoors to prevent the spread of radioactive dust. For crushing radioactive concrete to pieces up to two centimeters in size, it is possible to use jaw and cone crushers, which must also be automated and operate indoors. Further, we can refer to the experience of NASA in preparing for the colonization of the Moon, where it is planned to extract oxygen from regolith for rocket fuel and personnel breathing, and iron, aluminum and silicon - for industrial purposes at lunar stations [50]. The lunar regolith contains the highest percentage of oxygen and silicon – 16.4% silicon and up to 60.9% oxygen (9.4% aluminum, 2.3% iron) [51]. NASA plans to use the furnaces - Molten Regolith Electrolysis (MRE) reactors [50], in which the loaded regolith is melted with a high current (up to 850 A) and gaseous oxygen is separated at the anode, and melted metals and metalloids are separated at the cathode by direct electrolysis. The capacity of one such furnace is 3 tons of oxygen per year. The efficiency of these furnaces is almost 100%. There are no data in the literature on the distribution of radionuclides by components of direct electrolysis in MRE reactor; it is possible that the distribution will be the same as in metal smelting. Such a distribution of radionuclides by smelting products depends on the material being processed, the recycling technology, the applied temperature and the radionuclide itself [15], [16]. It can be assumed that radioactive cesium will be evenly distributed between gaseous oxygen and melt, and almost all of the radioactive cobalt will remain in the melt.

Radioactive oxygen obtained from concrete can be used to produce silicon dioxide. The technique of heating silicon in the presence of oxygen at a temperature between 600°C and 1200°C, used in microelectronics in the manufacture of microcircuits, is possible to be used to obtain silicon dioxide from silicon and oxygen obtained from concrete [52], [53]. Oxygen deactivation by sorbents can allow the use of oxygen in industry without restrictions. If the radioactivity of oxygen is such that it falls under any category of RW, then we can assume the possibility of using radioactive oxygen as rocket fuel on interplanetary flight trajectories. Nanopowders from radioactive silicon and silicon dioxide, for use in space microelectronics, can be obtained in the standard way, as for conventional nanopowders, for example, by laser ablation in an inert gas flow [54], evaporation under the influence of an electron beam [55]. One installation for the production of silicon nanopowders can produce up to 1 kg of powder per hour [54], silicon dioxide nanopowders - up to 7 kg per hour [55].

6. Recycling of radioactive metal structures

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The most promising method for recycling radioactive metals is their melting in a new generation of plasma-arc furnaces with preliminary surface decontamination in an annealing furnace [21]. In this case, the bulk of the radioactive component passes into the slag, and the radioactive residue is evenly distributed throughout the entire volume of the melt. The metal deactivated in this way can be used in industry without restrictions. Slags form about 10% of the initial mass of scrap [20]. Slag processing includes, for example, drying, grinding in a ball mill, roasting, and final grinding to obtain powders of various degrees of dispersion [24]. Plasma-arc furnaces allow melting 12 tons of radioactive metal per hour.

One of the most productive methods for obtaining iron nanopowders is the jet-levitation method, which makes it possible to obtain up to 1 kg of iron nanopowder per day [56]. Radioactive iron nanopowders can be used to manufacture parts and structural elements of space probes and planetary rovers by the same additive and powder methods that are used in modern metallurgy. Although there is no data on how radioactive powders behave under various conditions - elevated temperatures and pressures used in additive technologies.

7. Economic benefit

Below we can see the price of various nano- and micropowders [57] that can be made from radioactive material. Their sale should partially cover the costs of RW storage and processing:

The price of 1 kg of nanopowders (Euro): **Si** – 24 - 237, **SiO_2** – 17 - 42, **Fe** – 24 - 155, **Fe_3O_4** – 17 - 54, **Al** – 59 - 400, **Al_2O_3** – 16 - 195, **CaO** - 36, **AlSi** – 128, **FeSi** – 128, **C** – 8 – 73, **SiC** – 29 - 59.

The price of 1 kg of micropowders (Euro): **Si** – 16 - 55, **SiO_2** – 28, **Fe** – 18 - 34, **Fe_3O_4** – 4 - 6, **Al** – 15 - 98, **Al_2O_3** – 19 - 41, **CaO** – 14 -36, **C** – 19 – 47, **SiC** – 5 - 47.

The world consumption of nanopowders by the aerospace industry is 270 million euros per year (2% of the total world industrial consumption; the main consumers are metallurgy, electronics and medicine), which corresponds to the mass of nanopowder consumed 4500 tons per year at an average price of 60 Euro per 1 kg (data for 2019) [58]. That is, in the future, the aerospace industry can become an alternative consumer of radioactive waste recycled into nanopowders of metals, silicon and their oxides. However, to date, there are no scientific data on the properties of radioactive nanopowders and their application in industry.

8. Conclusion

From the above reviews on the recycling of radioactive scrap metal and concrete, industrial processing of slag from metal remelting, ash from the incineration of radioactive waste and industrial waste, it can be concluded that modern technologies make it possible to recycle any radioactive waste in order to obtain useful material from them for use in the aerospace industry. Taking into account the fact that the expansion of mankind into space is inevitable and the expansion of lunar and Martian missions, as well as missions to satellites of large planets, is planned, it can be assumed that any radioactive waste in any amount will be recycled in order to use them as active elements of spacecraft, which will solve the sore problem of radioactive waste disposal in terrestrial conditions.

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