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REVIEW OF NUCLEAR WASTE TREATMENT METHODS

Aditya Kalamdani*, Jishnu Madabhushi*, Abhinav Tyagi* & Dr. Anita Kumari**
* Student, Thadomal Shahani Engineering College, Chemical Engineering Department, Bandra West, Mumbai, Maharashtra
** Professor, Chemical Engineering Department, Thadomal Shahani Engineering College, Bandra West, Mumbai, Maharashtra

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Abstract:

Nuclear energy is being generated at an exceptionally higher rate now than what it was 50 years ago. The world is coming to realise that nuclear energy is our answer to the energy crisis we have been facing. It is clean, it is green and it does not give off polluting gases during its generation. However, the waste that is left behind after a batch of nuclear fuel has lost its potency, is quite harmful to both people and the environment as it is radioactive. We need to look at ways to treat this nuclear waste so that it doesn't negatively affect the environment. Broadly, there are multiple ways to deal with this waste and it is difficult to elaborate on all of them, but a few ones have been touched on in this paper and talked about regarding their process and their futures.

Introduction:

There are many uses of radioactive materials which improve or facilitate human activities or the quality of life of people. These uses are given in different fields of technology, ranging from power generation to supply entire cities or areas, to medical and industrial uses, even the smoke detectors in buildings. All these applications generate radioactive waste that may represent risks to the environment or to human beings, but it is necessary to have special attention to the management of radioactive waste.

Nuclear waste must be treated properly to make it safe for disposal. This includes its collection and sorting; reducing its volume and changing its chemical and physical composition, such as concentrating liquid waste; and finally, its conditioning so it is immobilised and packaged before storage and disposal.

In this paper, we review and discuss popular chemical treatment methods of radioactive waste categorising them in two categories: i) Liquid waste treatment methods and ii) Solid waste treatment method. In addition, there are information about the current state of research and technologies which have been proposed for the treatment of radioactive waste.

Treatment Methods of Nuclear Waste:

Liquid Type:

(i) ION Exchange Method:

Ion exchange methods have extensive applications to remove soluble radionuclides from liquid waste produced in nuclear fuel cycle operations, radioisotope production, and research facilities. It is very effective in transferring the radioactive content of a large volume of liquid into a small volume of solid.

The Ion Exchange/Sorption method is used for aqueous liquid type waste. In the early days of the nuclear industry, the use of ion exchange processes in water and wastewater treatment was well developed. Various recent studies have been devoted to the preparation and investigation of new ion exchangers that can face some of the difficulties associated with the industrial application of this treatment option and improve its performance. Examples of these tested new ion exchangers are composite materials that consist of one or more sorbents combined with another material. The composite can be inorganic or organic and can be a sorbent. The produced material is granular with sufficient strength for column use from sorbents that do not form or only form weak granules. This part will deal with the operation of the ion exchange method and its application in the nuclear industry.

The process involves the exchange of ionic species between a liquid solution and a solid matrix containing ionizable polar groups. When the exchangers are fully loaded, they are decommissioned and disposed of as radioactive waste. Alternatively, many organic ion exchange materials can be regenerated with strong acids or bases to produce radioactive liquid waste with high salt content and activity. Ion exchange media are available in many combinations of natural or synthetic, organic or inorganic materials, and in cation or anion exchange forms. Many media are also available in a variety of physical (eg, ball or powder) and chemical forms (eg, H^+ or Na^+ counterions). Cation exchangers are used to remove cationic species such as Cs^+ or Sr^{2+}, while anion exchangers remove anionic species such as Cl^-. Highly specific ion exchange media are readily available for radionuclides common in the nuclear industry, such as Cs and Sr, but these are frequently encountered in non-nuclear power applications. Ion exchange processes can be operated in batch or continuous mode. In a batch operation, a measured amount of medium is added to a tank of liquid, mixed and allowed to equilibrate for a period of time. The medium can then be filtered from the liquid. This type of operation is suitable for small-scale desktop operations. In continuous operation, the medium is contained in a vessel and the liquid is pumped through the medium under pressure [1].

A wide variety of materials are available for ion exchange of radioactive liquids. These materials are available in a variety of forms, have very different chemical and physical properties, and can be naturally occurring or synthetic. Natural mineral compounds such as clays (eg, bentonite, kaolinite, and illite), vermiculite, and zeolites (eg, analcite, chabazite, sodalite, and clinoptilolite) exhibit ion exchange properties. Natural zeolites were the first materials to be used in ion exchange processes. In 1985, British Nuclear Fuels PLC (BNFL) successfully commissioned an ion-exchange effluent plant (SIXEP) that uses naturally occurring clinoptilolite to remove cesium and strontium from fuel cooling tank water [2]. A large number of organic materials also exhibit ion exchange properties; these include polysaccharides (such as cellulose, algal acid, straw and peat), proteins (such as casein, keratin and collagen) and carbonaceous materials (such as charcoal, lignite and coal). Nowadays, clay materials and zeolites are replaced by synthetic materials.

Synthetic ion exchangers are produced by creating chemical compounds with the desired physical and chemical properties. They can be inorganic (mineral) or organic (generally polymeric) based. Some of the important synthetic inorganic ion exchangers are zeolites, titanates and silikitanates, transition metal hexacyanoferrates, while some of the organic counterparts are polystyrene divinylbenzene, phenol, acrylic [2]. Composite ion exchangers consist of one or more ion exchangers combined with another material, which may be inorganic or organic and may itself be an ion exchanger. The reason for producing a composite material is to produce a granular material with sufficient strength for use on an ion exchanger column that does not form itself or only forms weak granules [2].

(ii) Acid Digestion Method:

Acid digestion is a method of treating organic liquid type radioactive waste. Acid digestion is a technology for the oxidative destruction of some liquid organic wastes (hexane, Tributyl Phosphate) and organic components of mixed waste such as cellulose (paper), polyethylene, latex rubber, Tyvek, neoprene, polyvinyl chloride, polystyrene ion exchange resins, filters, plastics, and/or organic chlorinated cutting oils that can reduce waste volume 20 to 100 times.

Acid digestion refers to a chemical oxidation process to convert organic wastes into gaseous wastes and stable solid residues. The main reactions of the acid decomposition process for the treatment of radioactive waste can be shown using four reactions:

$$C_mH_n + n/2H_2SO_4 \rightarrow nH_2O + n/2SO_2 + mC \quad (1)$$

$$C + 2H_2SO_4 \rightarrow 2H_2O + 2SO_2 + CO_2 \quad (2)$$

$$3C + 4HNO_3 \rightarrow 4NO + 2H_2O + 3CO_2 \quad (3)$$

$$5C + 4HNO_3 \rightarrow 2N_2 + 2H_2O + 5CO_2 \quad (4)$$

Sulfuric acid serves both to carbonise the waste material and to oxidise it to CO, as shown by reactions 1 and 2. However, reaction 2 is somewhat slow, and nitric acid serves as a better oxidant according to reactions 3 and 4. The yield of plutonium can be estimated by laboratory tests that indicate that plutonium is readily converted to slightly soluble plutonium sulphate in the fermenter and remains largely as a precipitate with the rest of the process.

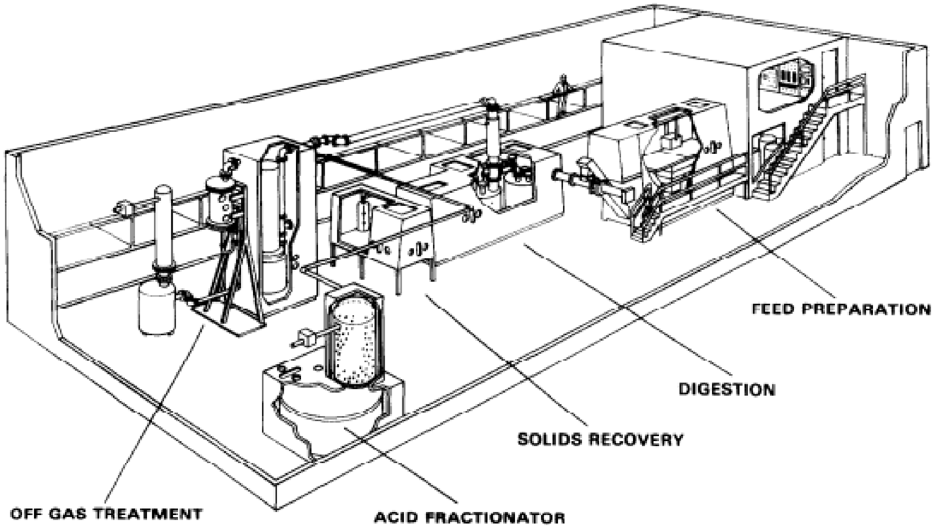

Figure 1: Radioactive Acid Digestion Test Unit (RADTU) (United States of America) [3].

The residue produced during the acid digestion of combustible waste materials consists mainly of sulphates and oxides of inert "filler" materials in the waste itself. The residue accumulates in the fermenter acid and must be separated from the acid by filtration or sulfuric acid distillation. The resulting dried residue has a similar volume to that resulting from the direct combustion of combustible waste.

The chemical safety aspects of the acid digestion process have been extensively investigated with regard to any possible formation of internal organic compounds or pressurisation of the system. The tests conclude that nitration reactions occurring in the process are highly unlikely. The degree of nitration depends on the reaction temperature and the concentration ratio of sulfuric acid to nitric acid, and decreases rapidly at both high temperature and high sulfuric acid to nitric acid ratios (eg >8) due to attack by organic materials. For the acid digestion process, the high ratio of sulfuric acid to nitric acid (greater than 20) along with normal operating temperatures of 250°C causes rapid oxidation of organics rather than nitration. Because few organonitrites are stable above 200 ^{O}C. they are not expected to accumulate in the system. The possibility of nitration reactions occurring in acid condensate from acid operations was also investigated and found to be highly unlikely. The nuclear safety aspects of acid digestion have generally been addressed by various countries through the use of critical-friendly designs. Independent studies and analyses have been conducted to identify the minimum critical amount of fissile material that can be processed in individual systems.

Some of the advantages of the acid digestion method are that the chemical reactions are easily controlled by adjusting the amount of waste and nitric acid or the temperature of the decomposition, the process also takes place at a low temperature, which allows plutonium to be retained in a water-soluble, non-refractory form. The disadvantage is that the speed of the process is low and the construction materials are limited [3].

Solid Type

(i) Vitrification:

Modern nuclear fuel recycling, coupled with recent advances in nuclear waste vitrification, can help solve the nuclear waste problems facing the world today. Advanced fuel recycling technologies are able to effectively remove and convert the most radioactive parts of long-lived nuclear fuel (UNF). The primary resulting waste stream contains fission products, inert chemicals and trace amounts of transuranics. This waste stream can be effectively immobilised by vitrification of nuclear waste. Waste glass can be safely stored, transported and disposed of once suitable facilities are available.

Great progress has been made in increasing throughput and efficiency in waste vitrification facilities, as well as loading waste into waste jars. However, further improvements are likely through continued technology development activities. Among the most promising approaches to technological development are the development of: Melting technology and glass composition capable of tolerating significant amounts of conductive solids (e.g. Melter). Glassmaking recipes and approaches to melter operation that prevent pooling of molten material of salts due to increased concentrations of S, Cr, Cl, F and Mo in the waste. Canisters and storage systems that can dispose of high temperature wastes.Glass preparations that can prevent coarse crystallisation of phases that can reduce the chemical durability of the resulting waste form. Glass-ceramic wastes are shaped and processed to allow higher loadings of Mo, Zr, Th, Al and Fe in the wastes. Programs to investigate the use of other groups of glasses (eg phosphates) to improve the filling and properties of waste glasses. Methods of removing and reusing precious metals and other fuel components to reduce the amount of waste destined for disposal [4].

Although nuclear waste glass technologists are no longer on a steep learning curve, they will continue to contribute to the expansion of glass technology. Advances are likely to come in at least some of the following areas of intensive effort. Glass redox. Methods are being developed to reliably predict the reducing power of melt feeds, as well as means to monitor the redox state of the glass once it is formed. The development of reliable, inexpensive instrumentation for Mössbauer spectroscopy appears to be key here. Several advanced processing techniques, such as the application of expert systems and sol-gel technology, are carefully evaluated. Using expert systems in conjunction with remote technology already developed for the Defense Waste Processing Facility (DWPF) can lead to a much more reliable glass manufacturing process. Long-term testing should lead to further improvement in the quantitative prediction of glass properties [5].

(ii) Incineration:

The process of nuclear combustion aims to reduce the long-term hazards of long-lived actinides and fission waste products generated in the nuclear power cycles of fission reactors. The plan is to transmute these long-lived products by nuclear reactions to produce less dangerous stable or short-lived products; however, care must be taken to ensure that more hazardous materials are not produced and that any increase in short-term risks from the process does not outweigh the long-term benefits. For the actinides, the fission process is usually considered the most suitable reaction, since the number of resulting fission products is only a small addition to those produced during normal reactor operation. Transmutation by neutrons, charged particles, and y-rays has been studied, but mainly neutrons have been considered. A wide variety of neutron sources have been investigated, from fission and fusion reactors to accelerators and even nuclear weapons. Until recently, however, most of the more detailed studies used fission reactors.

The half-life should be as short as possible so that the material burns quickly. However, if too small, there may be heating problems due to the energy released during the transmutation process, and the isotopes may need to be diluted by incorporating them in some other material. If the lifetime is too long, then the amount of any isotope will build up in the nuclear combustion system until the rate of its production and destruction remains the same.

If nuclear combustion is to be developed as part of a waste management strategy for fission reactors, a number of technological challenges need to be addressed, the main ones being:

- improved methods for high-efficiency chemical separation of neptunium, americium, and curium and long-lived fission products from reprocessing plant effluents are needed
- improved methods must be developed to ensure that waste streams contain very low percentages of uranium and plutonium (as well as neptunium, americium and curium and long-lived fission products)
- it may be necessary to develop special reactors or other devices that produce high fluxes of neutrons or other particles and rapidly transmute problematic nuclides
- fuel and fuel cells that contain nuclides must be developed.
- a total fuel cycle integrating special reactors or other equipment with the rest of the conventional nuclear fuel cycle must be developed and analysed to determine the amount of radioactivity returned to humans and thus quantify the reduction in long-term danger from nuclear burn-up[6].

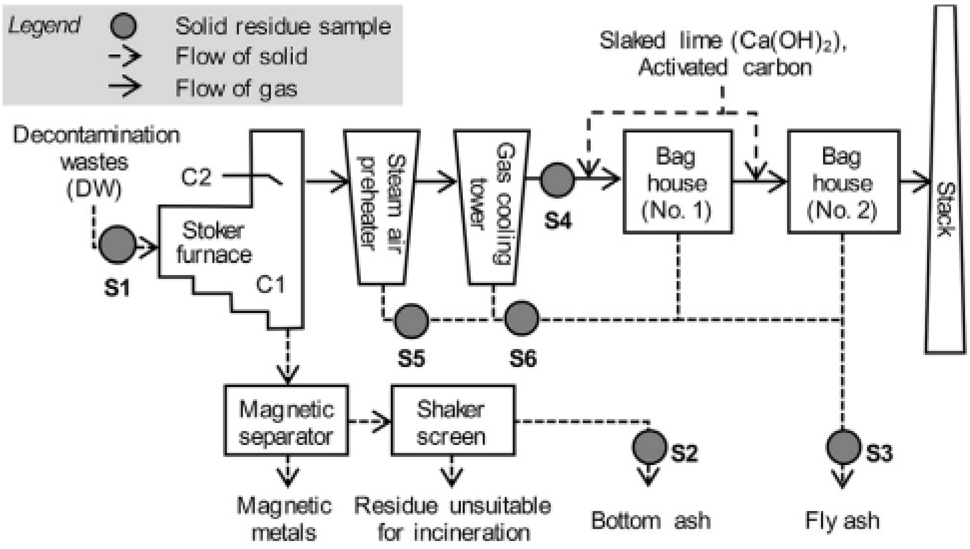

Figure 2: Schematic diagram of the incineration plant. The stoker furnace has a primary combustion chamber (C1) and a secondary combustion chamber (C2). Sampling points: S1, decontamination wastes in the stock yard; S2, bottom ash; S3, fly ash; S4, fly ash before the addition of slaked lime and activated carbon; S5, steam air preheater ash; S6, gas cooling tower ash [7].

Incineration in Micro Fission Chambers:

Combustion of transuranic elements by neutron-induced fission is a very promising way to reduce the long-term radiotoxicity of nuclear waste. The aim of Mini-Inca is to outline ideal physical conditions for the transmutation of minor actinides, especially 241–243Am, 237Np and 244–245 Cm [1]. For some actinides, there are large discrepancies in the neutron cross sections taken from the various nuclear data libraries evaluated. These cross-sections will play a dominant role in transmutation systems. For example, a factor of 20 has been pointed out for the 242 gAm thermal neutron capture cross section [2] from the JEF-2.2 (5500 b) and ENDF-B/VI (250 b) libraries. Computer simulations can lead to controversial results depending on the nuclear data library used. For an experimental approach to the burning rate of minor actinides and to provide a clear experimental reference, fission microchambers are of great interest.

The operating condition of the fission chamber, i.e. saturation current and operating voltage, is determined by four factors: the gap, specifically the difference between the cathode and anode radii, the mass of the deposit, and the nature and pressure of the gas filling. The goal is to find a compromise between these factors to define the best operating conditions of the fission chamber in the HFR neutron spectrum[8].

Conclusion:

In correspondence to the growth of energy demands across the world, nuclear energy and nuclear power generation sites have simultaneously grown considerably. With this development of the nuclear energy industry, the number of nuclear power plants has doubled, and several nuclear power plants have been retired.

The nuclear waste generated by them which is the main focus is naturally increasing. New treatment methods are being researched to tackle the problem of nuclear waste and ensure safe disposal.

Four important chemical treatment methods have been described in this paper for the treatment of solid and aqueous nuclear waste containing radioactivity. Different countries use modified versions of a particular treatment method to treat its radioactive waste. Waste containing long lived radionuclides must be treated, conditioned, stored and disposed of at a repository specifically designed for this purpose. Since radionuclides have decay rates ranging from days to thousands of years, proper segregation of wastes depending on their half-lives, and separate treatment and conditioning of these wastes, is an important factor in the overall scheme of radioactive waste management.

The general objective of waste treatment methods is to decontaminate nuclear waste to such an extent that the decontaminated bulk volume of waste can be either released to the environment or recycled. Waste concentrate is subject to further conditioning, storage, and disposal. Because nuclear power plants generate almost all categories of nuclear waste, nearly all processes are applied to treat radioactive effluents.

Although these processes and technologies are sufficient for effective management of radioactive waste at nuclear power plants, further improvements in this technology are still possible and desirable. The increasing cost of radioactive waste disposal provides an incentive to adopt procedures and techniques to minimise waste quantities and to develop new techniques to minimise volumes at the treatment and conditioning step.

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