STORAGE OF RADIOACTIVE MATERIALS

Güngör Gündüz
Kimya Mühendisliği Bölümü Orta Doğu Teknik Üniversitesi, Ankara 06531, Turkey

Keywords: Energy, storage, radioactive, waste, fission, vitrification, cementation

Contents

Summary

Radioactive materials are (i) naturally found in nature, (ii) produced in nuclear reactors for industrial use, and (iii) produced from the fission of nuclear reactor fuel. They may have very short or very long half-lives depending on the level of instability of the atomic nucleus. A radioactive substance is a continuously energy emitting system.

The volume of low-level solid radioactive wastes can be reduced by mechanical means. The liquid wastes can be chemically treated to precipitate and concentrate the waste. Dilute solutions can be also treated by ion exchange. Physical treatments such as evaporation and filtration accompany chemical treatment.

The separation of fission products from other elements can be carried out through a series of operations, decladding, dissolution, and extraction. As a result, three major components, depleted uranium, plutonium, and fission products are separated from one another.

The radioactive chemicals can be calcined and then vitrified to end up with an insoluble product. It can then be cemented for final storage. Cementation is done in noncorrosive tanks, which can be buried in underground tunnels if the level of radioactivity is high. Tanks containing low level cemented radioactivity can be stored in isolated open fields. Noble gases are diluted and discarded to atmosphere.

1. Introduction

Radioactive materials and radioactive wastes are energetic systems and continuously emit energy in the form of radiation. The radiation emitted is absorbed in several different forms by the target materials. The initial energy is usually degraded into heat energy. The storage of either radioactive materials or radioactive wastes needs special consideration for the personnel working with them, for the people sitting close by the source of radioactivity, and the environment.

The methods of handling, protection, and processing of all types of radioactive sources have been shortly described in a concise form in this article. The emphasis has been given to the wastes discharged from nuclear reactors.

2. Radioactivity and Fission

Nuclear radiation is the emission of massive or non-massive particles from the unstable nucleus of an atom. The massive particles are alpha (α), and beta (β) particle while gamma (γ) particle is non-massive and has the structure of a photon. These particles are usually highly energetic and cause physical, chemical, and biological changes when absorbed by the target materials. Therefore, radioactive materials can be considered as sources that continuously emit energies.

Alpha and beta radiations have short ranges because they carry electrical charges and easily interact with the atoms on the pathway of propagation, and they cause ionization. However, gamma radiation is in the form of photon and it can propagate over long distances.

It can be absorbed by a target material through different reaction mechanisms known as, (i) photoelectric absorption, (ii) Compton scattering, and (iii) pair production. The collision probability of a gamma photon with the target material increases with the atomic number of a nucleus, therefore, the absorption of a gamma photon by a nucleus through one of the above reactions increases as the nucleus gets heavier.

This is why small size radioactive materials are carried and stored in lead boxes or capsules. Lead has an atomic number of 82 and is known to be the best material for small size radioactive sources.

Each radioactive material has a half-life that denotes the time needed for the decay of half of the radioactive nuclei. The half-life may change from a fraction of microsecond to billions of years depending on the structure of the nucleus. There exist several naturally occurring radioactive elements or isotopes such as carbon-14, potassium-40, vanadium-50, rubidium-87, cadmium-113, cerium-142, etc.

Thorium and uranium are two heavy elements occurring on the earth and they have no stable isotopes. Uranium (U) exists in two isotopic forms, one is U-238 and the other is U-235. The former has a natural abundance of 99.27 %, and the latter 0.73 %. U-235 can be fissioned with slow (thermal) neutrons to produce nuclear energy, and it is called a fissile material. U-238 can be fissioned with very energetic neutrons, and it is said to be a fissionable material. In nuclear power reactors, nuclear energy is obtained from U-235. In fission U-235 nucleus is split into two new nuclei according to the following reaction.

$$U^{235} + n^1 \rightarrow X + Y + 2.43\,n + \text{energy}$$

The neutron absorbed by U-235 destroys its metastability and splits it into two new nuclei X and Y, meanwhile, 2.43 neutrons are emitted. This number is a statistical average of neutrons emitted from all fission reactions. The missing mass between the left and right hand sides of the above equation is converted into energy according to Einstein's well-known formula $E = mc^2$.

Most of the X and Y materials produced in fission have an atomic mass number range between 70-170 and they are called fission products. The mass spectrum of fission products is a bimodal curve having a double hump like an Asian camel. The fission yield is high around atomic mass numbers of 95 and 140, and low around 115, and also falls rapidly towards 70 and 170 on either side. The major concerns of radioactive wastes are all about the fission products, some of which have high radioactivity.

The reaction probability of a neutron with U-235 nucleus increases as the neutron energy is lowered. Therefore, the fast neutrons produced must be slowed down to increase fission rate in a nuclear reactor. For this purpose ordinary light water, heavy water, or graphite are used in nuclear reactors. However, these materials partly change into radioactive isotopes such as tritium (H-3) or carbon-14 during the operation of a reactor.

In fission chain reaction only one neutron is consumed in every step, and 1.43 neutrons out of 2.43 neutrons produced in each reaction, must be consumed somewhere to have a steady state operation of the reactor. Most of these extra neutrons are absorbed by control rods and structural materials in the reactor.

Some of them escape and they are stopped in the concrete wall, the so-called biological shield around the reactor. Very few of them cannot be stopped and they go to the environment. So, in summary, we can say that the radioactivity in nuclear plants is due to (i) mostly fission products in the fuel, (ii) moderator which slows down neutrons, (iii) structural materials, and (iv) shielding walls.

In order to improve the burn-up in the reactor, U-235 content is enriched from 0.73 percent to 3-5 %. Although most of the U-235 is burnt, some small quantities still remain at the end of a reactor cycle.

Another important reaction takes place between U-238 isotope and slow neutrons. As mentioned earlier, U-238 is not fissioned with slow (i.e. thermal) neutrons, but it captures a neutron and changes into U-239, which, then undergoes beta decay twice, yielding first neptunium-239 (Np^{239}), and then, plutonium-239 (Pu^{239}) as shown below.

$$U^{238} + n^{1} \rightarrow U^{239} \xrightarrow{\beta^{-}} Np^{239} \xrightarrow{\beta^{-}} Pu^{239}$$

Pu-239 is also a fissile material, and it is synthetically produced in nuclear reactors. It also fissions and gives nuclear energy.

Thorium (Th) exists only in one isotopic form (i.e. Th-232) in nature, and it is also a fissionable material. In addition, it also interacts with thermal neutrons and gives Th-233, which undergoes beta decay, first gives protactinium-233 (Pa-233), and then uranium-233 (U-233), as shown below.

$$Th^{232} + n^{1} \rightarrow Th^{233} \xrightarrow{\beta^{-}} Pa^{233} \xrightarrow{\beta^{-}} U^{233}$$

U-233 is also a fissile material and contributes to the energy production in the nuclear reactors.

The radioactivity artificially produced does not come out only from nuclear fuels. Significant amounts of radioactive materials are produced in commercial and research reactors. They are used in medicine to treat tumors or used for radioactive diagnosis. Radioactive materials are used in the industry for a number of miscellaneous tracing and measuring purposes such as the measurement of level, thickness, moisture, shape (i.e.

radiography) etc. Each radioactive source must be handled properly obeying certain regulations for the safety of the people.

The management and storage of radioactive materials has been a major concern for the public and for scientists to minimize the risks of contamination and the exposure of people. The experience of technologists showed that the radioactivity can be managed and controlled, and the most important problem is now localized on the long-term storage of high level radioactive wastes.

On-land nuclear weapon experiments, A-bomb disaster in Japan, and accidents in the former Soviet Union in Ural area and in Chernobyl taught a great deal about what to do and what not to do.

The level of radioactivity is measured in terms of the radiations emitted per unit time and also in terms of the total energy content and the biological damage caused in the tissue.

The radiation emitted from one transition in nuclear decay is called one Becquerel (Bq). The radiation damage in the tissue depends on the type and the energy of individual particle also. The combined effect is expressed in terms of Sievert (Sv) [1].

Radioactive wastes can be found in solid, liquid and gas states, and are classified into three groups:

- Low level waste (LLW) (of order ~10^4 Bq/l)
- Intermediate level waste (ILW) (of order ~10^7 Bq/l)
- High level waste (HLW) (of order ~10^{10} Bq/l)

The classification is based on the level of radioactivity in a unit volume, and not on the total volume or weight.

LLW can be handled without shielding. ILW needs shielding for the protection of the personnel. HLW needs both shielding, and cooling because of the heat generated due to high radiation.

3. Storage of Fuel

Nuclear fuels have very low activities since their half-lives are extremely long. However, large amounts of fuels emit radiation that needs precaution. The very specific point that separates fuel storage from wastes is the nuclear phenomenon called "spontaneous fission". Heavy fissile nuclides can split into two without interacting with a neutron [2]. The fission caused without a neutron is called spontaneous fission.

The spontaneously fissioned nucleus releases neutrons, which, in turn, interact with fissile nuclei and accelerate fission reactions. Although the half-life for spontaneous fission is very small (i.e. 5.5×10^{15} years for U-238), its effect can not be ignored. The only way to eliminate chain reactions is to allow sufficient space between fuel bundles so that any neutron generated by spontaneous fission can decay into a proton without starting a fission reaction. The half-life of the neutron (e.g. 10.6 min) as well as the mass and geometry of fuel are the parameters used in the calculation of the placement of fuels in the storage room [3].

4. Transportation of Radioactive Materials

The transportation of radioactive materials is subject to international regulations. They are usually carried in lead shielded containers. The activity on the surface of package can not be more than the allowed limits that depend on the type and energy of radiation. The radiation on the surface of a package can not exceed 2 mSv/h, and it can not exceed 0.1 mSv/h at a distance of one meter away from the source. In exceptional use the limit is 10 mSv/h [4]. Each package has to be labeled with the sign of radioactivity.

5. Volume Reduction of Low level Wastes

Liquid wastes can be stored in specially designed tanks after concentration operation, but must be changed into solid form for long term storage [5-7]. Solid wastes can be put into three categories; (i) compressible or combustible materials which make up about 70 % wastes; the polymeric materials, paper and cloth or other combustible materials are all included in this group, (ii) hard materials, such as metals, coatings and linings that need fragmentation for size reduction, (iii) wastes originating from plant conversion and operations.

5.1. Mechanical Volume Reduction

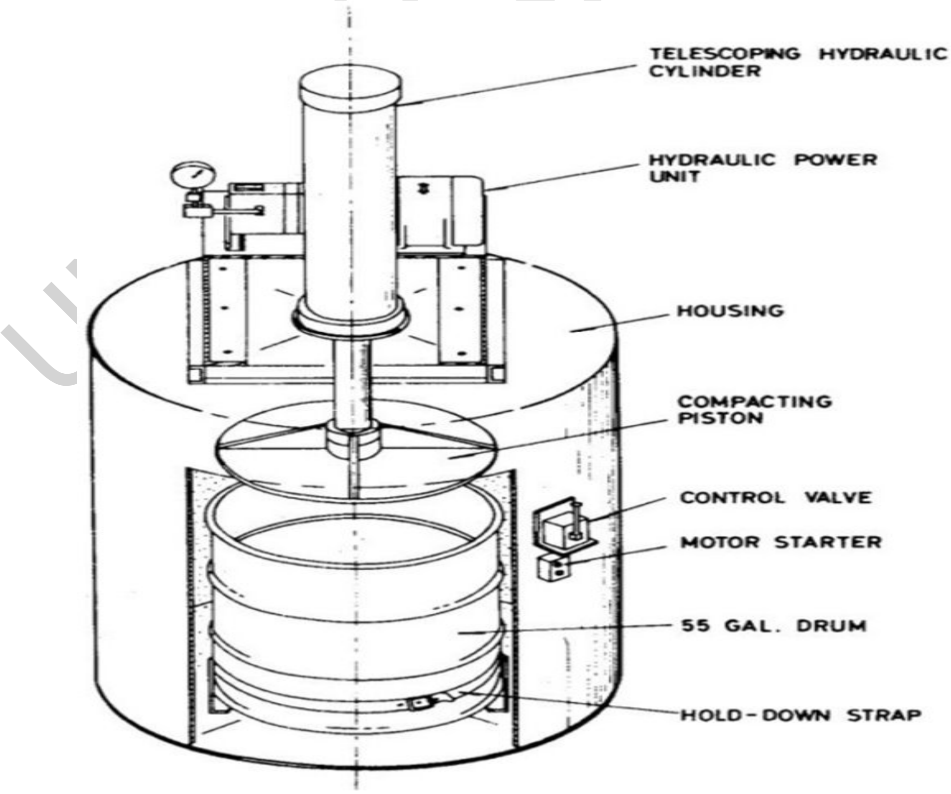

Figure 1. A compacting unit for solid wastes (Reproduced from Ref.8 with the kind permission of International Atomic energy Agency)

Compacting and fragmentation can reduce the volume of low level solid radioactive wastes. Low-density materials such as plastics, papers, glassware and some metallic items or boxes can be easily compacted by using a heavy-duty press. A 2-15 fold size reduction can be accomplished depending on the type of material and its initial form.Wastes can be directly dumped into the compacting mould. For small-scale compaction a force of 10 tons may be sufficient. For large objects hydraulic presses working at 400 tons may be used. A small-scale compacting unit is shown in Fig. 1.

5.2. Fragmentation

In nuclear plants, miscellaneous large items have to be dismantled and fragmented, because it is difficult to carry them due to their large size, such as water tanks, compressors, ventilation hoods, heat exchangers, ovens, etc. Size reduction is done by using shearing machine, guillotine, saws, electric arc and oxy-acetylene torch cutters.

5.3. Treatment of Liquid Wastes

Liquid radioactive wastes are first collected in small tanks, carried to the site for concentration, treatment, incineration, and final storage. The collecting tanks and the auxiliaries must be made of corrosion resistant materials, and the joints must be sufficiently tight to prevent any leakage. Commonly used corrosion resistant materials are stainless steel and polyethylene. Glass lined and ceramic tanks are also corrosion resistant but they are fragile, so they are not used much. The container must be placed into a shield for further safety. Figure 2 shows the picture of such a container.

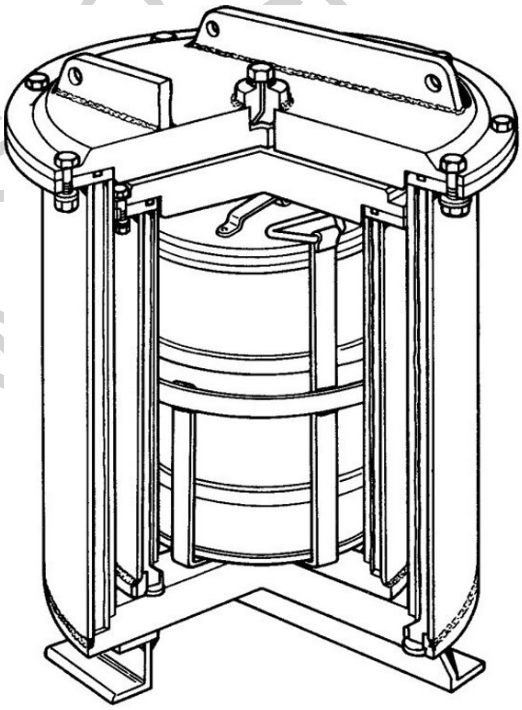

Figure 2. A shield container (Reproduced from Ref.7 with the kind permission of International Atomic energy Agency)

Small volume wastes produced intermittently can be segregated into storage tanks at the basement of laboratories, or outside the buildings and collected in hold up tanks. The tanks are at least two in number and they are charged periodically. The waste running into the first tank is kept there until the tank gets full, and then the second tank is put into service. After the relatively short-lived isotopes sufficiently decayed, the content of the first tank is carried away for further processing.

-
-
-

Bibliography

1. United Nations, (1993) *Sources and Effects of Ionizing Radiation.*
2. J. R. Lamarsh, (1966) *Introduction to Nuclear Reactor Theory*, Addison-Wesley Publishing Co.
3. A. F. Thomas, F. Abbey, (1973) *Calculational Methods for Interacting Arrays of Fissile Materials*, Pergamon Press
4. International Atomic Energy Agency, (1996) Regulations for the Safe Transport of Radioactive Materials, No. ST-l.
5. International Atomic Energy Agency, (1972) Storage Tanks for Liquid Radioactive Wastes: Their Design and Use, Technical Reports, no: 135.
6. International Atomic Energy Agency, (1968) Design and Operation of Evaporators for Radioactive wastes, Technical Reports, no: 87.
7. International Atomic Energy Agency, (1964) Technology of Radioactive Waste Management Avoiding Environmental Disposal, Technical Reports, no: 27.
8. International Atomic Energy Agency, (1970) The Volume Reduction of Low-Activity Solid Wastes, Technical Reports, no: 106
9. A. Kahraman, (1997) Development of Carrier Usage for the Treatment of Radioactive Effluent, in Treatment Technologies For Low and Intermediate Level Waste from Nuclear Applications, Tecdoc-929, International Atomic Energy Agency, pp. 33-50, pp. 111-121
10. International Atomic Energy Agency, (1968) Chemical Treatment of Radioactive Wastes, Technical Reports, no: 89.
11. International Atomic Energy Agency, (1967) Operation and Control of Ion-Exchange Process For Treatment of Radioactive Wastes, Technical Reports, no: 78.
12. F. Shengtao, G. Li, C. Li, W. Benli, W. Likong, (1997) Radiobiological Waste Treatment-Ashing Treatment and Immobilization with Cement, in Treatment Technologies For Low and Intermediate Level Waste from Nuclear Applications, Tecdoc-929, International Atomic Energy Agency, pp. 33-50
13. International Atomic Energy Agency, (1970) Bituminization of Radioactive Wastes, Technical Reports, no: 116.
14. C. J. Wood, (1995) Dose Reduction Methods: Chemical and Physical, in Radiation Protection at Nuclear Reactors, Editor: C. J. Malteskos, Medical Physics Publishing, pp. 353-396

15. A. D. Johnson, P. R. Maul, and F. H. Passant, (1990)Radioactive Waste Storage and Disposal in the UK, in Advances in Nuclear Science and Technology, Editors: J. Lewins and M. Becker, Vol: 21, Plenum Press, pp. 255-296

16. J. Prawitz and J. Rydberg, Acta. Chem. Scand. (1958), 12, 385

17. M. Benedict, T. H. Pigfort, (1957) *Nuclear Chemical Engineering*, Mc-Graw Hill Book Co.

18. M. Benedict, T. H. Pigfort, and H. W. Levi, (1981) *Nuclear Chemical Engineering*, Mc-Graw Hill Book Co.

19. J. P. Olivier, (1975) The Management of Fission Products and Long-Lived Alpha Wastes, in Advances in Nuclear Technology, editors: E. J. Henley and J. Lewins, Vol: 8, Academic Press, pp. 141-172

20. R. Roy, (1982) Radioactive Waste Disposal, Vol: 1, The Waste Packaged, Pergamon Press.

21. G. Gündüz, B. Yeter, P. Tuğlu and S. I. Ahmed, J. Mater. Sci. "(1981), 16, 221-225

22. H. F. Aly, M. R. El-Sourougy, (1997) Prospects for Storage and Disposal of Low and Intermediate Level Radioactive Waste in Egypt, in Planning and Operation of Low Level Waste Disposal Facilities, International Atomic Energy Agency, pp. 101-121

23. R. L. Andersen, (1995) Criteria for Radiological Releases, in Radiation Protection at Nuclear Reactors, editor: C. J. Maletskos, Medical Physics Publishing, Wisconsin, pp. 21-44

24. P. H. Carrigan, ()Hydrologic and Hydrodynamic Aspects of Low Level Radioactive Waste Disposal to Streams, Proceedings of the Symposium on Low Level Waste Disposal, Site Characterization and Monitoring, Vol: 2, Editor: M. G. Yalcintas, NUREG/CP-0028, CONF-820674, pp. 273-294

25. F. L. Parker, ()Effect of Releases from Near-Surface Low level Waste Sites on Surface Water Quality, Proceedings of the Symposium on Low Level Waste Disposal, Site Characterization and Monitoring, Vol: 2, Editor: M. G. Yalcintas, NUREG/CP-0028, CONF-820674, pp. 295-327

26. International Atomic Energy Agency, (1999) Topical Issues in Nuclear, Radiation and Radioactive Waste Safety.

27. International Atomic Energy Agency, (1995) *Environmental Impact of Radioactive Releases*.

28. I. Uslu and O. Aliefendioglu, (1997) Evaluation of NSARS-Test Case 2C and Lessons Learned for its Application to Turkish LLW Sites, in Sixth International Conference on Radioactive Waste Management Environmental Remediation, ICEM 97, editors: R. Baker, S. Slate and G. Benda, The American Society of Mechanical Engineers, pp. 623-626

29. R. Baker, S. Slate and G. Benda, editors(1997): Sixth International Conference on Radioactive Waste Management Environmental Remediation, ICEM 97, The American Society of Mechanical Engineers.

30. International Atomic Energy Agency, (1987) In Site Experiments for Disposal Radioactive Wastes in Deep Geological Formations, Tecdoc-446.

31. Y. Sakabe, (1997) Design Concept and its Development for the Rokkasho Low Level Radioactive Waste Disposal Center, in Planning and Operation of Low Level Waste Disposal Facilities, International Atomic Energy Agency, pp. 123-132

32. H. Bohm, K. D. Closs, and K. Kuhn, (1981) Radioactive Waste Disposal, in Advances in Nuclear Science and Technology, editors: J. Lewins and M. Becker, Vol: 13, Plenum Press. pp.1-72

33. J. S. Carlsson, F. Karlsson, and P. Riggare, (1997) Assessment of the Long Term Performance of the Swedish Final Repository for Radioactive Waste, in Planning and Operation of Low Level Waste Disposal Facilities, International Atomic Energy Agency, pp. 353-368

Biographical Sketch

Güngör Gündüz graduated from the Chemical Engineering Department of Orta Doğu Teknik Üniversitesi, Ankara, Turkey with B.Sc.(1968) and M.Sc.(1970) degrees. He got his Ph.D. from the Nuclear Engineering Department of Iowa state University, USA, in 1975. He then joined the Chemical

Engineering Department of Orta Doğu Teknik Üniversitesi as a faculty member. He has numerous publications on nuclear shielding, in-core fuel management, urania-gadolinia fuels, boron nitride and boron carbide coated nuclear fuels. Currently, he is doing research on advanced ceramics, polymers, and surface coating.