RADIOACTIVE WASTE MANAGEMENT IN THE USSR EXPERIENCE AND PERSPECTIVE

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ABSTRACT

Both radioactive waste decontamination and the improvement of nuclear power safety are key problems in the development of nuclear power in the USSR. The new political situation in the USSR, the publication of previously classified data on the development of the nuclear power industry, which has gained a wide public response, and the serious aftereffects of the Chernobyl accident have led to the revision of the strategy of the development of nuclear power as well as the experience in radioactive waste management.

Radioactive wastes are produced by nuclear fuel cycle facilities, nuclear power plants, naval nuclear power plants, research nuclear reactors, medical scientific centers, and industrial enterprises, which use ionizing radiation sources in their activities.

The problem of radioactive waste decontamination in the USSR is acute and, to a great extent, it depends upon radioactive waste buildup at the initial stage of the operation of nuclear industry enterprises.

The radiochemical industry in the USSR came into existence in 1949 when a radiochemical plant was put into operation in the South Urals in the Chelyabinsk region to extract plutonium from natural uranium irradiated in military uranium-graphite reactors. Later the plant was converted, firstly, for reprocessing spent fuel of naval transportable power reactors and then of the Soviet light water reactors WWER-440. Today the plant ensures all of the demands in spent fuel reprocessing of such reactors constructed in the USSR and abroad.

A new radiochemical plant will be built for reprocessing spent fuel of light water reactors WWER-1000 spent fuel assemblies are transported there for storage.

The largest quantity of different radioactive wastes from reprocessing of irradiated uranium has been accumulated on the site of the first radiochemical plant in the South Urals. Within a 40-year period of radiochemical plant operation, the reprocessing technology has changed substantially, several times. Firstly, the regeneration technology was based on precipitation processes. The process of uranium and plutonium purification from fission products and their separation was developed at the Radium Institute (Leningrad) under the guidance of Academician Khlopin V. G. The technology was based on slightly soluble sodium uranyl acetate ($NaUO_2(CH_3COO)_3$) precipitation from nitric acid solutions of irradiated uranium. Plutonium, being in the six valence state in the form of sodium plutonyl acetate, coprecipitates isomorphically with $NaUO_2(CH_3COO)_3$ or remains in the solution, if it is reduced to plutonium (4) or plutonium (3).

In the first case, uranium and plutonium purification from fission products was achieved and in the second one, their separation.

In fact, during the first years of the operation of the radiochemical plant, acetate-nitrate solutions made up the bulk of high-level radioactive wastes with sodium nitrate concentrations exceeding 100 g/l and sodium acetate concentrations making up 60-80 g/l. These solutions occupied a large volume and their storage presented great difficulties. However, it was impossible to achieve their considerable concentration by evaporation due to high salinity. Besides, the solutions contained a large quantity of a deficient reagent, sodium acetate. To reprocess acetate-nitrate radioactive wastes, a precipitation-crystallization-sorption technology was developed by the Physical Chemistry Institute of the USSR Academy of Sciences under the guidance of Academician Spits in V. I., which provided for the solution of the following three problems:

- Radionuclide concentration by precipitation of insoluble compounds with high sorption properties in relation to radionuclides-fission products;
- Production of high purity crystalline sodium nitrate, when it is not referred to the category of radioactive wastes and can be used as a usual substance, e.g. as a fertilizer or for producing alkali;
- Recovery of acetate-ion for its recycling in irradiated uranium reprocessing technological processes.

Radionuclide concentration was achieved by their coprecipitation with such low-soluble compounds as iron and chromium hydroxides, iron and nickel sulfides and nickel ferrocyanide.

Radioruthenium and radiostrontium are concentrated on nickel and chromium hydroxides, zirconium, niobium and protactinium radionuclides on iron and nickel sulfides, and cesium radionuclides are coprecipitated with nickel ferrocyanide.

The concentrate of fission products prepared in the form of a suspension was supplied to a long-term storage,

and the clarified solution after acidification by nitric acid was concentrated by evaporation. Simultaneously, acetate acid was distilled and caught in a plate column, sprayed with alkali. From distillation residue containing 1,100-1,150 g/l sodium nitrate its crystallization and even recrystallization were realized, if it was required for higher purification. The technology ensured concentration of fission products in a volume approximately 100 times smaller as compared with the volume of the initial solutions.

Subsequently, as a result of the plant's reconstruction, the precipitating technology of irradiated uranium reprocessing was replaced by ion-exchange technology and then by liquid extraction with the use of a tributyl-phosphate solution in an inert diluent as an extractant. Salinity of high-level radioactive wastes (water-tail solutions) decreased several times. As a result, there was no necessity in the precipitation-crystallization-sorption technology of high-active waste reprocessing. The waste reprocessing technology was reduced to evaporation with nitric acid recovery for its recycling in the technological process of irradiated uranium regeneration and preparation of radionuclide concentrate (distillation residue).

The variety of fuel compositions reprocessed, as well as changes in regeneration technology, caused the accumulation at the plant of radioactive wastes with various radioactive levels and considerably different chemical compositions.

The radiochemical plant came on stream when just technological processes of plutonium extraction from irradiated uranium had been basically mastered. The problems of safe waste handling, staff health and environmental protection had not been investigated sufficiently. High-level wastes were supplied for storage in stainless steel cooled tanks set in concrete canyons, also lined with stainless steel to protect the environment from radioactive contamination in case of corrosion.

But in the very first years of the plant's operation from 1949 to 1952, the radioactive wastes were partially discharged into an open water reservoir situated near the plant. I'll speak about it later.

Storage of liquid high-level waste solutions and suspensions is of potential danger. High specific activity of solutions causes their self-heating and radiation-chemical transformations of their organic and inorganic components. As a result, dangerously explosive gases and corrosive substances are formed, as well as decomposition of acids and formation of precipitates capable of absorbing radionuclides, causing local overheated areas. In a case of the violation of technological standards, accidents are possible.

In 1957, because of the failure of a cooling system of a tank with high-activity nitrate-acetate wastes and the resulting self-heating up to dried salt and increasing temperatures up to 300-350 degrees Centigrade, a chemical explosion and dispersion of radioactive fission products took place.

There were no victims, but a part of the territory of the region was contaminated. The effective urgent measures taken such as evacuation of the population from the nearby areas, decontamination of the territories, made it possible to avoid serious aftereffects for the personnel and population. Nevertheless, some measures had to be taken to develop reliable and safe methods of radioactive waste storage.

The storage facilities for high-level liquid waste were reconstructed. New, more reliable tanks were built. Additional measures were taken to increase the reliability and safety of liquid high-level waste storage, such as:

- Continuous air blowing over the surface of the solution to diminish the concentration of explosive gases (hydrogen, methane), generated from the high-active solutions;
- Maintaining a strictly, controlled temperature in every tank;
- Achieving a possibility for remote pumping of liquid waste from one tank into another in case of temperature rises;
- Continuous control and correction of acidity to eliminate solid phase precipitation, etc.

In the middle of the 50's the USSR, as well as other nuclear countries, began to develop techniques for transforming liquid radioactive wastes into a solid state with reliable radionuclide fixation in stable matrices, suitable for long-temperature safe storage. Preference was given to preparation of glass-like materials. Developments of methods of high-level liquid waste vitrification in the USSR was carried out in two directions. The first direction was a two-stage vitrification with waste calcination at the first stage. Various flowsheets of calcination processes, including a boiling layer apparatus, a spluttering calcinator, a rotating horizontal calcinator were investigated. The second direction, which received a wider development, is the so-called single-stage method of preparing phosphate and borosilicate glass-like materials in a ceramic melter without preliminary calcination. Dehydration, calcination of wastes and their melting with fluxing additions are conducted in one apparatus, where liquid high-level wastes and fluxing agents are dosed directly. For obtaining phosphate glass, the orthophosphoric acid is dosed as a fluxing agent and for borosilicate glass the boron-containing mineral datolite is dosed. The heating of glass-like melt is carried out by conducting alternating current through the glass melt.

Despite the bulky technological flowsheet, the technique of single-stage vitrification is characterized by high

capacity and allows the high alkali metal salt-containing wastes to be processed.

The 500 l/h vitrification facility for liquid high-active solutions was put into operation at the plant in 1987 after almost 10 years of testing were carried out at the 100 l/h facility using model solutions. The process is based on radionuclide introduction into phosphate glass, prepared in a ceramic melter made of high-alumina zirconium refractory material with molybdenum electrodes. Vitrified wastes are poured through special drains into 0.2 m^3 canisters. After cooling, three of such canisters are placed into metal containers with a diameter 0.63 and 3.4 m high. Glass blocks placed into metal containers are put into surface storage, equipped with a forced system of air cooling and with a powerful gas-purification system. Permanent temperature and gas control of the containers will be carried out during the whole period of storage (20-30 years). About 1,000 m^3 high-active solutions with 3.9×10^6 Ci total activity were treated at the vitrification facility during 1987-1988. Total weight of the obtained glass blocks constitutes 160 t. In 1988 the vitrification facility was shut down. It is currently under reconstruction. When reconstruction of the facility is completed, plans are to carry out combined vitrification of high-level and medium-level radioactive solutions. In this case, medium-level radioactive solutions will play the role of a fluxing addition.

Liquid radioactive wastes with specific activity of 10^{-5}-1 Ci/l refer to the medium-level radioactive waste category. These wastes include decontaminating solutions, off-gas system condensates, solutions after extractant washing, etc.

Such wastes were temporarily collected in Lake Karachaj, which is situated not far from the reprocessing plant. In the first years of operation some part of technological wastes with an activity of 120×10^6 Ci was discharged into the lake. At present, about 95% of the total radioactivity has been sorbed in the deposits of the lake. The area of the lake prior to the waste discharge was 0.26 km^2, the water volume was 143,500 m^3.

Because of a great potential danger of open storage of radioactive waste caused by the penetration of contaminated solutions into underground waters and the possibility of radioactivity, being spread by winds, the work on its covering with soil and subsequent reliable isolation has been carried out for the last several years. Work on the liquidation of the lake is to be completed by 1995.

The up-to-date technology of radionuclide concentration from medium-level radioactive solutions is based on separate concentration by evaporation of high and low saline solutions. Distillation residue solutions from evaporation of low saline solutions are intended to be used as a fluxing addition in the process of high-level waste vitrification. Distillation residues from evaporation of high saline solutions will be transformed into bitumen compounds and stored in shallow earth repositories. Condensates from the evaporation of the solutions are reprocessed in facilities for low-active waste purification. The low-active wastes are purified on ion-exchange resins. Purified water is supplied to the system which recirculates the water supply, and solutions from ion-exchange resin regeneration are reprocessed together with medium-active solutions.

Alongside solidification of liquid radioactive wastes, the USSR has been carrying out work on pumping such wastes down deep intaking formations for a long time. The work was already begun in the 1950's both in the Soviet Union and USA. The technique of underground disposal of liquid radioactive wastes consists of their controlled disposal into deep aquifers isolated from other formations and day surface with a system of injection wells. When assessing geological structures and intaking formations, it is mandatory to rule out radioactive wastes release into rivers, seas, lakes and earth surface, as well as the contamination of underground water fit for domestic and drinking purposes, mineral water and mineral deposits. In the 1960's, geological and hydrological, physico-chemical, thermal and health studies allowed to develop several projects for the underground pumping of liquid radioactive wastes. To avoid the accumulation of radioactive wastes on the surface, for several years the USSR, as a forced measure, had to pump liquid radioactive wastes down the zone of a very hindered water exchange between platform-type artesian aquifers, containing highly mineralized water and located outside an active tectonic zone. The natural rate of stratal water filtration in these aquifers constitutes several meters per year. The period of water exchange in such aquifers is commensurable with geological epochs. A factor such as radionuclide sorption in soils enhances the reliability of their retention. Soviet experts have repeatedly reported their experience in the underground disposal of liquid radioactive wastes at international conferences and symposia. The waste diffusion contours are monitored thoroughly. In spite of a satisfactory health and radiological situation around the test sites, lack of effects on the surface and shallow underground waters and lack of any dose commitment of the population, our present knowledge does not allow consideration of the technique of underground pumping of liquid radioactive wastes to be absolutely reliable. Therefore, this technique is unlikely to be used in the future.

Low- and intermediate-level wastes are produced as a result of NPP operation. They represent solutions after the decontamination of equipment, pipelines, premises, overalls and protective clothing; leaks from coolant equipment and loops; solutions after the flushing of ion-exchange resins and filtering materials. Liquid radioactive wastes are evaporated at the Soviet NPPs. The condensate is recycled into the circulating water supply system after decontamination and purification using coal and pearlite filters and ion-exchange resins, while the bottoms, containing 200-300

grams of salt per liter and $3.7x10^6$-$3.7x10^7$ Bq/l, as well as filtering and sorbitizing materials (their activity amounts to $3.7x10^6$-$3.7x10^9$ Bq/l) are stored temporarily in 400-5,000 m^3 separate tanks. The bottoms, filtering and sorbitizing materials must be transformed into bitumen and cement compounds. Such installations are being built at all NPPs. The first installation for bitumenizing liquid radioactive wastes of NPPs was built at the Leningrad NPP and put into operation in 1986. Its output is 500 l/h. Bitumen compound is pumped into and stored in bulk-type repositories. An installation for cementing liquid radioactive wastes is being built at the Novo-Voronezh NPP. After concentration and solidification, all wastes produced during the NPP operation are stored on NPP sites. Nuclear district heating plants will be an exclusion and their wastes are to be shipped from the site to the nearest regional radioactive waste disposal point. Solid radioactive wastes of nuclear power plants are stored in special concrete storage facilities. Inflammable solid wastes are subject to incineration and other wastes will be compacted to reduce their volume.

Radioactive wastes, produced as a result of the application of radionuclide sources in medicine, research and industry, are collected by regional plants responsible for the collection, processing and disposal of radioactive wastes. There are more than 30 regional points of such type in the Soviet Union. The Moscow Research and Production Association RADON executes the methodological guidance of the regional points of radioactive waste disposal.

A critical analysis of our previous experience in radioactive waste management and the necessity to recover certain territories, contaminated in the past as a result of insufficiently elaborated and sometimes forced technical approaches, have led to the development of the State Concept of radioactive waste management. It was devised taking into account the domestic and international experience.

Radioactive waste management technology must prevent harmful effects of radionuclides on the biosphere and, therefore, release of radioactive wastes of all types in environment is inadmissible. Spent nuclear fuel is not classified as waste in the Soviet Union. It must be reprocessed at radiochemical plants to extract unburned uranium and plutonium. Nuclear fuel of the first Soviet power reactors, perhaps, will be an exclusion, because its reprocessing may be almost unfeasible and economically unadvantageous. All high-level wastes must be transformed into solid forms stable for long-term storage (glass, mineral-like materials). Intermediate-level solutions, which do not contain long-lived radionuclides and transplutonium elements, must be transformed into bitumen, cement and polymeric compounds. Solidified high-level wastes must be disposed into stable geological formations below the zone of active water exchange to such a depth, which excludes a possibility of radionuclide release into the human environment.

As to the ultimate goal of radioactive waste management, i.e. their reliable isolation, it is not only their specific activity, but also the time for which they preserve their properties which are hazardous for human beings and environment, that are of principal importance. Therefore, the time period required for the decay of radionuclides in radioactive wastes down to a safe level is a governing factor in selecting a technique for their disposal and type of natural and engineered barrier for isolation. Isolation of long-lived radionuclides and transplutonium elements is most complex. Such radionuclides must be concentrated in a minimum volume, since their storage requires continuous monitoring and the possibility of removal, if a danger of their migration into the biosphere arises.

The Radium Institute in the USSR and Czechoslovak experts have developed a technique of high-level waste fractionation to isolate the most hazardous radionuclides into separate fractions. This technique is based on the utilization of an extracting agent, which belongs to the carbolide-class extractants (solution of cesium salt of cobalt decarbolide in a mixture of a polar solvent and polyethylene glycol). The technological flowsheet ensures the extraction of over 99% of cesium, strontium, transplutonium elements, and rare earth elements into separate fractions. A high-capacity installation is not required for the solidification of long-lived radionuclide fractions. The fractionation of high-level wastes can expand a choice of matrix compositions for the solidification of each fraction.

The USSR carries out investigations in different rocks, which may host a storage facility. A possibility of the construction of such repository directly on the site of the first radiochemical plant is being studied. It is of extreme importance, because it may exclude the transportation of large quantities of radioactive wastes.

Solidified low- and intermediate-level wastes, which do not contain long-lived radionuclides and transplutonium elements, may be stored in shallow land repositories with a reliable isolation, preventing possible migration of radionuclides. Radioactive waste repositories should be located in non-seismic areas, which show no promise from the national economy viewpoint.

The problem of reliable isolation of radioactive wastes requires joint efforts of the international scientific community due to the global nature of their potential hazards for mankind and, therefore, it should be solved via the widest international cooperation.

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