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## MANAGEMENT OF RADIOACTIVE WASTES AND COUNTERMEASURES OF MODIFIED STABILIZATION PROCESS

The paper presents a short review of a radioactive waste management, i.e. disposal practices for gases, liquids and solids discharged from nuclear power plants. In order to weaken a radioactive fallout into environment, several countermeasures, including natural zeolites, are taken. The article is concluded with some comments on the Balkan syndrome and on the energy production from the uranium fission reactions which in many countries of the world still presents more than 50% of the whole energy balance.

### 1. INTRODUCTION

The nuclear industry nowadays is able to provide information on the probable consequences of environmental contamination extended over a lifetime and to make the risk estimations.

The situation differs in most other industries. Discharge of potentially noxious materials into environment involves some risk, which may or may not be measurable. Usually the cheapest procedure is to discharge the waste in some way that will ensure a sufficient dilution to make it innocuous [1], [2].

If we decide on a particular waste management system or to license a particular kind of a power station, a much more deliberate weighing of costs versus benefit must be undertaken. One benefit of nuclear power is the difference between death and injury among uranium miners and those employed in processing industry and the corresponding figure for equivalent energy produced by the coal mining industry [3].

One can only dream about finding a true cost to benefit ratio, and the people responsible for approving a waste management system or a new power station are therefore faced in the last analysis with a value judgement, which is at least to some extent subjective. It is not a scientific decision. In the fullest sense, the decision is political.

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## 2. SOURCES OF RADIOACTIVE WASTES

Apart from the usual hazards associated with hardrock mining, the workers in uranium mines are exposed to radon and the decay products of the radium-containing ore. The large volume of air used for mine ventilation is ejected at high velocity from a stack which ensures its adequate dilution in the atmosphere [4].

The end product of the mill is uranium oxide. Radium originally present in the ore is one of the most toxic radionuclides. Various methods of treatment such as coprecipitation with barium render most of the radium insoluble.

The crude  $U_3O_8$  (70%) produced by the mills may be converted into metal  $UO_2$  or  $UF_6$ . The latter is used in separation of  $^{235}U$  from  $^{238}U$ . The content of  $^{235}U$  in natural uranium reaches 0.7%. The toxicity of natural uranium or  $^{238}U$  is such as that of toxic metal rather than of a radionuclide [5].

An operating reactor contains a very large inventory of fission products. A 500 MW (thermal) reactor after 80-day operation contains 400 000 000 Ci of fission products measured one day after shutdown [6]. This is equivalent to the activity of about 400 metric tonnes of radium. In an operating power reactor, the fuel is contained within a non-corrodible cladding – usually made of zirconium or stainless steel – and the fission products cannot get out unless the cladding is ruptured.

Fuel removed from the reactor is normally stored on sight for a considerable time to permit the decay of short-lived radioisotopes [7]. Storage facilities are usually deep tanks filled with water which acts simultaneously as coolant and radiation shield.

It should be stressed that by the end of this century the build-up of  $^{14}Cl$  in the atmosphere might become a significant source of radiation within the biosphere.  $^{85}Kr$  is liberated via fuel poisoning and by diffusion through fuel cladding (half-life of 10.4 years). In the reactors filled with ordinary water or  $D_2O$  coolant and in the presence of the moderator of heavy water, tritium is produced by neutron activation. Day-to-day mechanical failures can produce air-borne radioactive iodines [8]–[10].

Spent fuel after storage is dissolved in strong nitric acid, and both uranium and plutonium are extracted using an organic solvent. High-, medium- and low-level wastes are produced and left behind. The most difficult problem in the fuel processing industry is still very low-level liquid waste because it is produced in enormous volume [11].

Research centres, hospitals equipped with teletherapy machines, biological laboratories, isotope production plants, naval ships, ice breakers may be the sources of  $^{131}I$ ,  $^{32}P$ ,  $^{59}Fe$ ,  $^{51}Cr$ ,  $^{90}Sr$  and  $^{137}Cs$ .

### 2.1. ASSESSMENT OF THE CONSEQUENCES OF RADIOACTIVE FALLOUT AND APPLICATION OF COUNTERMEASURES

Thirteen years after the Chernobyl accident (April 26th, 1986) a great deal of mostly reliable information on its causes and consequences can facilitate the decision of how to minimize the probability of such a catastrophe and simultaneously of how

to find an optimal system of response countermeasures against incidental nuclear emergencies.

As a result of the accident, the environment was contaminated with radioactive materials of total activity of ca. 12 500 PBq. Today  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are considered to be the main contaminants. About 95% of the radioactive contaminants are deposited in the upper layers (5–8 cm) of undisturbed soil. The physicochemical analysis shows that the fallout is basically represented by the dispersed fuel particles as well as condensation- and adsorption-generated particles. The calculated inventory of a core at the moment of the accident was about 3600 PBq for the alkalies, with 330 PBq of cesium. The content of cesium released into the atmosphere varies from 15 to 40%.

The territory of Slovakia was contaminated by dry and wet deposits from the first Chernobyl cloud. In 1986, the average values of body surface contamination reached orders of  $\text{kBq/m}^2$  for  $^{137}\text{Cs}$ . The highest contamination was monitored in the eastern quarter of the city of Bratislava-Rusovce due to the strong rains in the first days of May [12]. The values of surface contamination by  $^{137}\text{Cs}$  reached there on average 8–10  $\text{kBq/m}^2$ . In 1986, the means for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  accumulated in the top layer of soil in the Slovak Republic were 2.18  $\text{Bq/m}^2$  and 0.145  $\text{kBq/m}^2$ , respectively.

At such a level of contamination only organized countermeasures were taken just at that time. Fifteen days after the radioactive fallout in the top layer of soil not thicker than 1 cm and 2 cm, the amount of  $^{137}\text{Cs}$  decreased to 96% and 4%, respectively. But one month later in the first centimeter of soil layer, there was 81% of  $^{137}\text{Cs}$ , while in the 1–2 cm and 2–3 cm thick layer – only 15% and 4% of  $^{137}\text{Cs}$ , respectively [13].

The half-life of  $^{137}\text{Cs}$  accumulated in 0–10 cm soil layer varies from 10 to 25 years. The cleanup of soil layer was slower in its upper part (24–27 years for the 0–5 cm layer) than in lower one (10–17 years). Usually, the processes of the cleanup proceed 1.2–3.0 times faster when soil is contaminated with  $^{90}\text{Sr}$  than when it is contaminated with  $^{137}\text{Cs}$ .

The uptake of contaminants present in soil by higher plants depends on several factors: soil characteristics and such biological characteristics of the plants exposed to radiation as: physiology, ecology and morphology, density, transpiration of plants, which influences the transport of elements from roots to leaves, and others. The coefficient of transfer of radionuclides to plants growing on sodded loamy soils of a podsol type is 1–3 times lower than the coefficient of transfer for sodded podsollic sandstone soils of a podsol type. Therefore, the improvement of decontamination of the sodded podsol as a result of the complex use of organic fertilizers, liming and high doses of potassium and phosphorus fertilizers reduces four-fold the content of  $^{137}\text{Cs}$  in crops, and when accompanied with water regime the improvement is 10 times lower.

It has been shown in practice that special agronomic, agrochemical and reclamation measures applied to contaminated agricultural lands permit us to decrease the cesium transfer from soil to crop by the factor of 1.4–4.0. Liming of acid soils decreases the above transfer by the factor of 1.4–1.7, while potassium fertilizers used in excess – by the factor of 1.7–2.5. Deep ploughing with digging combined with liming and potassium fertilizer allows a decrease in this transfer by the factor of 3–4.

Higher efficiency of natural zeolite in various industrial and agricultural applications is due to the fact that products made from zeolites preserve such main properties of these minerals as: high sorption capacity, ion exchange and dehydration–hydration abilities, thermal, acid as well as radiation resistance.

Domestic clinoptilolite from only one industrial deposit in eastern Slovakia, Nižný Hrabovec, exhibits a high selectivity towards  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . Zeolite mineral ore is characterized by an active component (clinoptilolite), whose content ranges from approximately 40 up to 80%. The uniformly exploited zeolite charge, i.e., zeolite of the identical properties, was applied both in simulation in laboratory and in field experiments to reduce (by currently occurring Cs (Sr)) the number of sites affected by radionuclides [14]–[16].

The laboratory tests were carried out in order to investigate the capacity of domestic clinoptilolite to adsorb some radionuclides, i.e.  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{65}\text{Zn}$ ,  $^{60}\text{Co}$ ,  $^{133}\text{Ba}$ ,  $^{125}\text{I}$ ,  $^{59}\text{Fe}$  in aqueous model solutions. The figure presents some typical adsorption characteristics of natural and  $\text{NH}_4^+$ -modified clinoptilolites, which were tested in aqueous solution of  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  labelled with  $^{65}\text{Zn}$  as a radioindicator. In the figure, we can find a mathematical description of adsorption process in terms of the Freundlich isotherm.

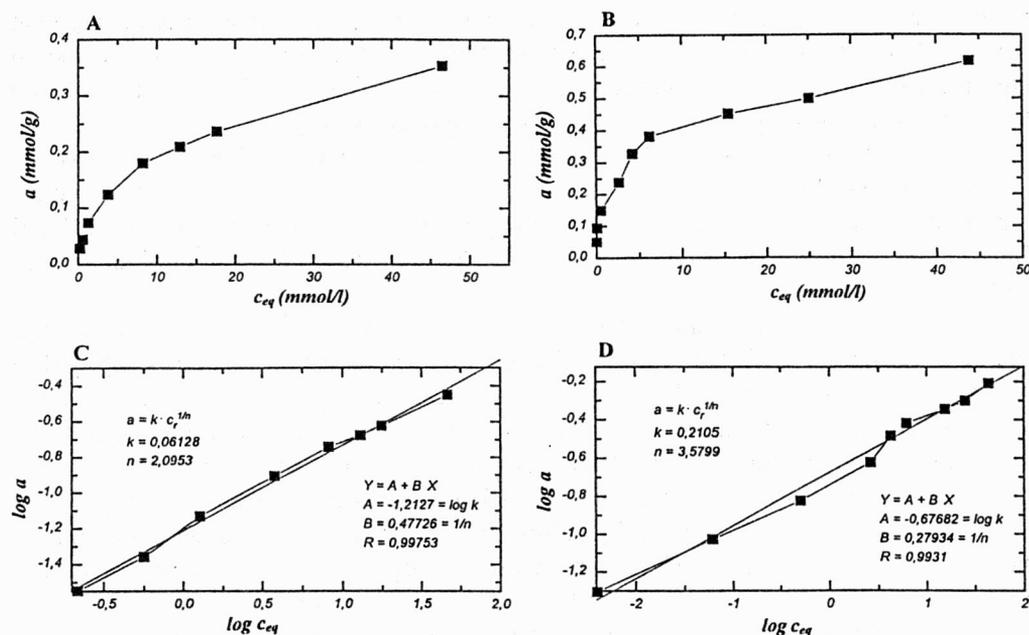
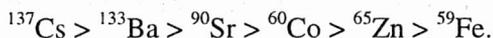


Figure. Adsorption isotherms in the systems: (A) aqueous solution of  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  – natural clinoptilolite, (B) aqueous solution of  $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ –  $\text{NH}_4^+$ -modified clinoptilolite, (C) linearized Freundlich isotherm representing case (A), (D) linearized Freundlich isotherm representing case (B);  $a$  – adsorption capacity [mmol/g],  $A$  – constant term by regression computation,  $\beta$  – regression coefficient,  $c_r$ ,  $c_{eq}$  – equilibrium concentrations [mmol/dm<sup>3</sup>],  $k$ ,  $n$  – constants of Freundlich equation of isotherm,  $R$  – correlation coefficient

To investigate the sequence of cations (radionuclides) selectivity towards clinoptilolite, numerous laboratory tests were performed. Some kinetic and thermodynamic data (i.e. chemical equilibrium constants, selectivity coefficients, diffusion coefficients) were calculated in order to verify the results. The following sequence of the mentioned radionuclides selectivity in aqueous solutions was confirmed [17]:



### 3. DISPOSAL PRACTICES

#### 3.1. GASES

Radioactive gases are mainly produced in reactors during spent fuel processing, isotope production and operation of research facilities. It is advisable to filter a contaminated air near to the source of its origin. Filters should be made of non-flammable material such as glass or asbestos fibres.

Small airborne particles are usually electrically charged. The particles are attracted to a surface carrying the oppositely charged electrostatic precipitants, from which they can be removed mechanically.

The most efficient air cleaning device other than "absolute" filters consists of a nozzle in which the air is mixed with steam. Then the mixture is expelled from a nozzle and enters an expansion chamber, where the steam condenses on the particles. After passing through expansion chamber, where the air is scrubbed with water jets, the removal efficiency for 0.3 micron particles approaches 99.9%.

Incinerator off-gases are cooled and some fly ash is removed by passing it through a cooling chamber fitted with baffles. After this stage a preliminary or bag filter is used followed, if necessary, by absolute or charcoal filters.

Radioactive iodine in molecular form is fairly easily absorbed in alkaline scrubbers by copper or silver mesh filters, but in the form of methyl iodide it can only be immobilized by an activated charcoal filter [18]. These filters have to be kept cool, which is indispensable not only for removing the decay heat of adsorbed halogens, but also for remarkably higher efficiency of  $^{85}\text{Kr}$  absorption on cold charcoal.

#### 3.2. LIQUIDS

In Denmark and Sweden, practically all liquid wastes are treated by evaporation and this method is also widely used in Japan. Residues that remain after evaporation may be mixed with cement, fused with glass frit or various ceramic mixtures or incorporated into melted bitumen. The product is then handled as a solid waste. Evaporation is the most straight-forward and apparently the simplest method of treating a radioactive liquid waste.

In most waste tanks, a sludge settles down. It may retain up to 90% of its activity and on a copious precipitate of metallic hydroxides up to 90% of wastes are adsorbed

[19]. After separation of these sludges further treatment of a clear effluent consists in the addition of lime and sodium carbonate. In some cases, ferric chloride, clay or other additives are introduced in carefully chosen steps of the process.

The effluent that is formed during a well-controlled flocculation process has a low content of total solids and after filtration (to remove traces of flocs) it can be passed through a cation exchanger suitable for the removal of radioactive contaminants [20]–[22].

One of the most promising ways to fix high-level waste is to incorporate it into a glass. This “self-heating” waste is too active to be treated in conventional way. Several methods have been developed for evaporation and subsequent calcination of wastes. The calcination is carried out in a heated steel container, a fluidized bed or a spray calciner. The waste mixed with glass-forming fluxes (borax, lead oxides) is heated to about 9000 °C.

The oil industry has developed methods for making fissures in rock in order to encourage movement of oil to gas. This process has been adapted to the disposal of medium-level wastes [23].

Due to the presence of salt in a deep salt mine the water has been absent there for millions of years and geological study proves that water cannot rapidly penetrate the salt bed. The excavated galleries of salt mines are large and stable tunnels, suitable for storage and roamy enough for safe work with active loads.

### 3.3. SOLIDS

Much of the solid material can be reduced in volume by incineration or baling under high pressure. In the latter case, bales and non-combustible waste are likely to be buried in sparsely populated regions. For example, in France quite low-level solid wastes are put into concrete containers which are then filled with cement mixture so that the end product is a large concrete block. These blocks are stored under a roof on a concrete floor. In Canada, on the other hand, similar wastes are put into open trenches at the Chalk River and covered with the local sandy soil.

A very effective conditioning process is fixation of solids with bitumen or asphalt. Bitumen is very resistant to radiation, impermeable to water, has a low melting point, and some mechanical flexibility. In general, bitumen begins to have the advantage over concrete as the method of choice of “fixing” otherwise mobile waste radionuclides.

The International Atomic Energy Agency has issued a code of practice in management of radioactive wastes by hospitals, research institutes and industry when no special facilities are available on the site [25].

## 4. CONCLUSIONS

Current nuclear age history has started since the middle of the Second World War, when Italian scientist Enrico Fermi prevailed on best-known scientist of all time Al-

bert Einstein to sign a letter to President Franklin D. Roosevelt indicating the importance of the importance of the uranium fission reaction to produce a tremendous explosive force. The United States government launched a massive research project for the study of atomic energy – designating the Manhattan Project.

Fermi and his group achieved the first sustained nuclear reaction on December 2nd, 1942 under the bleachers at Staag Field at the University of Chicago.

The first atomic bomb as the most dangerous environmental impact of nuclear fission was tested in the desert near Alamogordo, New Mexico, on July 16th, 1945. The heat from the explosion vaporized the 30 m steel tower on which it was placed and melted the sand for several hectares around the site.

The uranium atom bomb by nuclear fission splits in many different ways. In all, over 200 different fission products are produced, with half-lives varying from less than a second to more than a billion years (three of the most worrisome isotopes are  $^{90}\text{Sr}$ ,  $^{131}\text{I}$  and  $^{137}\text{Cs}$ ). In addition, the neutrons produced in the explosion act on molecules in the atmosphere to produce  $^{14}\text{C}$  and other radioisotopes.

About 10 tonnes of depleted uranium was lately released into environment during the Balkan war. The use of depleted uranium is not even known only from this region. So far, thirty soldiers, participants in peace forces in the Balkan, died of the so-called “Balkan Syndrome”, the illness possibly caused by depleted uranium in the weapons. However, all participants, including civil local citizens, were surely more or less affected by environmental disaster and danger caused by bombing of chemical plants, electric power stations, fertilizer and water purification plants, petroleum refineries, high-voltage transformers, etc.

Coming back to nuclear power stations, there are still many countries all over the world whose energy production by fission of uranium represents more than 50% of the whole energy balance (France, Canada, Slovakia, Sweden, Belgium, Japan, Lithuania).

However, the power of nuclear fission has not yet been harnessed in a sustained, controlled reaction of appreciable duration that produces more power than it consumes. Controlled nuclear fission processes would produce almost no radioactive waste products. Despite insurmountable technical problems involved in harnessing a fission energy, the promise of the abundant, relatively nonpolluting energy source makes its pursuit worth a massive effort.

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ZARZĄDZANIE ODPADAMI PROMIENIOTWÓRCZYMI I ŚRODKI  
PRZECIWDZIAŁAJĄCE ZMODYFIKOWANEMU PROCESOWI STABILIZACJI

Dokonano krótkiego przeglądu sposobów postępowania z odpadami promieniotwórczymi. Mogą to być sposoby ich usuwania z gazów, roztworów lub ciał stałych powstających w elektrowniach jądrowych. Aby zmniejszyć opad promieniotwórczy, podejmuje się różne środki zaradcze, w tym m.in. stosuje się naturalne zeolity. Artykuł kończą uwagi na temat tzw. syndromu bałkańskiego i wytwarzania energii w wyniku rozszczepiania uranu, dzięki czemu liczne państwa na świecie wytwarzają więcej niż 50% energii.

