TECHNIQUES AND TECHNOLOGIES OF DEFINITION OF THE CONTENTS OF URANIUM AND PLUTONIUM ISOTOPES IN RADIOACTIVE WASTE

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In this review, the issues of radiation and nucleus safety at dealing with nuclear waste (RW) are considered. The examples of already worked out technologies on definition of the isotopes contents of uranium and plutonium in radioactive waste are adduced; the possibilities of implementation of techniques with application of electron linac are shown.

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1. SOME ASPECTS ON THE HANDLING OF RADIOACTIVE WASTE

First of all, it is about safeguarding a radiation security. According to the Radiation Safety Standards and main sanitary regulations, the firm radioactive waste (RW) is considered to be radioactive in case its activity makes more than 10^{-8} Ci/kg [1]. With provision of specific activities for ^{238}U and ^{235}U it corresponds to concentrations of $3 \cdot 10^{-5}$ g/g and $4.7 \cdot 10^{-6}$ g/g. The liquid RW are considered to be radioactive, if the contents of some radionuclide or mixture of several radionuclides exceeds acceptable concentration levels established for water. So the liquid RW on value of a specific activity is subdivided into following categories:

– radioactive waste with low activity-is lower than 10^{-5} Ci/l;

– radioactive waste with mean activity-from 10^{-5} up to 1 Ci/l;

Secondly, the safeguarding of nuclear safety is the main issue in nuclear power engineering. In this direction the researches are conducted and the different aspects of radioactive waste handling and its environmental impact are discussed.

So, in article [2], the conditions, which may cause a self-sustaining chain reaction in a process of handling of the RW possessing small quantities of fission stuffs, are considered. The typical waste from plants producing HGA is analyzed as well as firms of a fuel cycle such as NPO "Radon", where the waste possessing fissile stuff is stored for long term and prepared for storage. Liquid waste with concentration of uranium of 1-5 Ci/l also refers to them. The similar situation can arise also at other plants of a fuel cycle. Even within low specific contents of fission stuffs in huge volumes, forming thousand cubic meters, the probability of originating of self-sustaining chain reaction of fission_is not expelled. The self-sustaining nuclear reaction became possible at the contents~3 g of ^{235}U on 1 kg.

As at process and storage the contents of fissile stuffs in waste can change (increase), the originating a self-sustaining nuclear fission reaction is possible in certain conditions. That is why a systematic analysis of nuclear safety of all RW handling system is indispensable, which is split into three phases:

– the analysis of a radioactive waste from the point of view of danger of originating of self-sustaining chain reaction of fission;

– the analysis of technological processes of conversion;

– the analysis of nuclear safety at long-time storage of processed waste;

All kinds of waste are divided into two groups by characteristic of material in order to define criterion of nuclear safety. The waste, containing mainly graphite belongs to the first group, the waste of the second one mainly contains hydrogenous moderators. In work [3] it is proposed to use surface density of fission stuffs as criterion of nuclear safety for storehouses. In Table 1 the surface density for different compounds of uranium and plutonium is adduced.

The analysis of nuclear safety of storehouses, in which barrels with a volume of 200 l, diameter of 560 mm and thickness of a steel wall of 1,5 mm are arranged closely in four circles by height, shows that in order to avoid the self-sustaining chain reaction of fission, each barrel should contain not more than 50 g of plutonium and up to 80 g ^{235}U.

These conditions are provided for firm waste with low activity NPO "Radon". However, the volume of waste decreases in 70…100 times during incineration and it can move over to category of the waste with mean activity.

Therefore, it is necessary to carry out analysis on the contents of fission stuffs before incineration of waste with low activity.

In case of liquid waste the safe concentration of aqueous solutions of uranium and plutonium makes 9 and 7 g/1 accordingly. However, there is a probability of fission stuffs sedimentation and obtaining of large concentration that also demands the analysis of their contents.

In work the typical waste is reviewed, which is supposed to be stored or to be sent for processing in NPO "Radon" and the calculations for definition of the secure specific contents ^{235}U with regard for content of water (Table 2) are adduced.

Besides the rate of fission stuff concentration, the chemical, mechanical and physical processes within which the fission stuffs are separated from nuclides absorbing neutrons are important in order to determine a probability fro originating of self-sustained fission chain reaction.

Table 1. *Secure and critical surface density of fission stuffs (g/см^2 ^{235}U or ^{239}Pu)*

Connection	Critical	Secure
$^{239}Pu + H_2O$	0,2	0,14
$U^{*} + H_2O$	0,37	0,26
$U^{**} + H_2O$	0,5	0,35
$^{239}Pu + C$	0,036	0,025
$U^{*} + C$	0,052	0,036
$U^{**} + C$	0,06	0,042

Note*: uranium is enriched 90 and 5% on ^{235}U accordingly*

Table 2. *The secure specific contents of fission stuffs for firm waste*

Waste	Elemental composition, mass lobe of components, %	Enrich-ment of uranium, %	Secure specific contents of waste, ^{235}Ug/кg
Gum	C 70-90 H 3-10 O 1-10 S 3-10	4,4 90	0,78 0,48
Graphite	C~100	4,4 90	0,09 0,08
Ceramic of boards	O 50-70 Si 20-30 Al 10-20 Fe 1-2	4,4 90	3,6 2,1
Polyethylene items	C 86 H 14	4,4	12
Fluoroplastic	F 41-76 C 21-24 H 0-7 Cl 0-30	90 4,4	13,5 7
Organic glass	C 60-80 H4 O 10-30	90 4,4 90	6,9 4,3 3,9
Zirconium waste	N 3 Zr 100	4,4 90	1,6 0,9
Plastic, tissue of PHV	C 38 H 5 Cl 57	4,4 90	38 8,1

In October 1999 the International conference TOPSEAL- 99, which was organized by Belgian and European nuclear companies at support of the American nuclear company, IAEA and Agencies on atomic engineering of countries OECD (NEA) was held in Antwerp (Belgium) [4]. The representatives from 22 European countries and also from Egypt, Russia, the USA, Taiwan, Southern Korea participated in it. The main subject of the conference was creation of international storehouse for the spent fissile material. Records of the conference contain short information on status of RW dumping for the last 10 years as well as the prospects of a research direction by treatment of the RW. The following activities are planned:

– formation of general approaches for treatment of the RW;

– working out of technical measures on environmental protection, safety and stable exploitation;

– working out of the project of storehouse for the long-lived RW, in particular, prolongation of activities aimed to solve existing technical problems and to coordinate technical and legal issues between the organizations - participants.

More than 50 projects on the RW treatment problems for the total amount of around €37 millions were completed in the period 1994-1998. These projects concerned mainly the following subjects:

– aspects of safety at RW dumping;

– field experiments on the underground exploratory installations;

– research of features of the main technological processes of RW treatment and other.

The field experiments were conducted in Belgium on three underground trial installations in different geological conditions and with the purpose of:

– check and demonstrating of the concepts of dumping;

– filling and seal of storehouse;

– monitoring for long-term condition a component of storehouse.

2. SITUATION IN UKRAINE

In Ukraine the problem of the spent fissile material treatment appeared in 1997, when the exportation of spent fissile to Russia for processing and dumping was stopped [5] (in 1997, only 4,6% from the existing amount of spent HGA accumulated on atomic power stations (APS) and research reactors was sent to Russia).

At Zaporozhye APS it is planned to create a storehouse of a dry type in connection with a critical situation on conventional storage of exhaust fissile material in basins of exposure.

The main producers of a radioactive waste in Ukraine and places of its concentration for today are:

- APS (spent fissile material);
- industry on a mining of uranium and process industry (65,5 mill. t of radioactive waste is accumulated);
- medical, scientific, industrial and other firms;
- the Ukrainian state association "Radon" (more than 5000 m^3 of radioactive waste is accumulated);
- the zone of disposal Chernobyl' APS (more than 1,1 bill. m^3 of a radioactive waste).

The main objects of accumulation of the greatest amount of the RW with high activity in Ukraine are the nuclear stations, on which its partial primary processing and temporary storage take place.

The radioactive waste at APS consists of liquid and firm waste. The liquid waste are formed as a result of disturbance of hermetic sealing of a primary loop, sinks per head, labs, regeneration waters of special water purifying, and at realization of decontamination activities. The product of processing of a liquid radioactive waste-"the bottom remainder» is stored as it is or it is dabbed in order to decrease volume by a method of evaporation. At all APS there is no full work cycle of primary processing of liquid radioactive waste. Only at Zaporozhye and Khmel'nitskiy APS the evaporation up to salt content of 1500…1600 g/l is carried out. At the rest of APS, liquid radioactive waste is stored as "distillation residue", which does not meet the requirements stipulated in the norms and rules of treatment of liquid radioactive waste.

The firm radioactive waste is formed as a result of maintenance and repair of generating sets. On January 1, 1998 in storehouses of APS in Ukraine, there were 261126 m^3 of a firm radioactive waste and 25216 m^3 of "distillation residues".

3. OBJECT "SHELTER"

Now fissile material is in object "Shelter" arranged in five modifications:

– pieces of an active zone;

– fuel as dust with the comparative sizes of fragments from 1 micron up to hundreds of microns;

– stuffs keeping fuel, which represent glassy mass formation in consequence of interaction of molten fuel with structural materials;

– solvable salts of uranium and plutonium, the concentration of uranium in which is 0,3 g/l;

– solvable compounds of uranium and plutonium, which has sedimented.

Isotopes of cesium, strontium, plutonium, uranium and americium represent the main radionuclides.

On weight the stuffs keeping fuel, in percentage terms make: uranium oxides up to 1…8%, potassium – 1-2%, calcium – 4…10%, magnesium – 3…7%, aluminum – 2…12%, ferry lactase – 3…10%, zirconium – 2…4%, silicon-up to 60% and more. The congestions of masses containing fuel in the "Shelter" present nuclear danger conditioned by a probability of origination of self-sustaining chain reaction. Under influencing of different causes and due to physical-chemical processes, the state and the properties of the masses containing fuel is changing which makes radiation safety situation at the "Shelter" worse. With the passage of time one may observe modification of the structure and properties of the masses containing fuel, which is conditioned:

– by appearance of cracks and decay of lava on large enough chunks;

– by destruction of lava with formation of radioactive dust;

– by interaction of alkali contained in water, with radionuclides and by formation of new solvable compounds.

4. METHOD FOR DETECTION OF THE CONTENTS OF TRANSURANICS IN RADIOACTIVE WASTE

In France in CEA-DRN-DER- SSAE and DGA-DRET-ETCA-DPN the technique for detection of the contents of transuranics with the help of a electrons linac is being worked out. The method founded on results from counting photofission-induced delayed neutrons from transuranics [6]. In activity, the linac of electrons with energy up to 30 MeV, mean current of the beam 140 μA, repetition frequency ranging from 6,25 up to 400 Hz and pulse width 2,5 μs was used. The main problem at debugging of the techniques was a neutron background, the life-time of which was about 20 ms. In this connection, the test-pieces of the RW were placed in protection from polyethylene and cadmium. Neutron detectors for a monitoring of photoneutrons and neutrons of fission were also placed here. In activity, there were 5 samples used: U-Pu with the contents of uranium of 3,83 g and plutonium of 1,9 g accordingly; samples of ^{238}U (weight 100, 194, 292 g;) sample of ^{235}U (weight 1,9 g). Samples were placed in cylindrical units from polyethylene with diameter of 10 cm and length of 20 cm and from glass with diameter of 10 cm both height of 9 cm and concrete with diameter of 56 cm and height of 110 cm with density 0.95, 1.70, 2.35 g/cm^3 accordingly. In Table 3 the minimum weights in grams are shown, which were registered in the given experiment.

Table 3

Element	Matrix		
	$(CH_2)n$	SiO_2	Concrete
UPu	0,024	0,003	0,029
^{235}U	0,016	0,027	0,058
^{238}U	0,052	0,052	-

The obtained values of minimum weights for matrixes U-Pu and ^{235}U, determining sensitivity of technique, correspond to concentrations $8{,}94\cdot10^{-7}$ and $1{,}77\cdot10^{-6}$ g/g.

5. TECHNOLOGIES

The company SGN and department of physical researches (DRP) of the French commission on an atomic energy (CEA) conduct intensive developments on creation of engineering of the active and passive analysis on definition of the contents of fissile stuffs in firm and fluid radioactive waste. The necessity of development of methods of the fissile analysis is conditioned by the following facts:

– low level of surface contamination in storehouses (some milligrams PuO_2);

– greater amount of containers, contents of which is necessary to analyze in short time;

– the development of technologies of a nondestructive examination is based on implementation of neutron-source ^{252}Cf and neutron generators [7].

"COSAC" SYSTEM FOR 20-LITER BAGS

This system is designed for measurement of waste with very low activity located in 20 l volumes with a mild matrix. The main principle of the given system is the activation by neutrons from ^{252}Cf of a source with the subsequent registration a gamma-quanta and neutrons of fission. The system "Cosac" includes a source and detectors, and also movable protection for a source (Fig. 1). Usage of gamma - detectors allows logging 1 mg of plutonium PWR during 600 s. Owing to the neutrons of fission the low limit of detecting makes 13 mg of plutonium PWR.

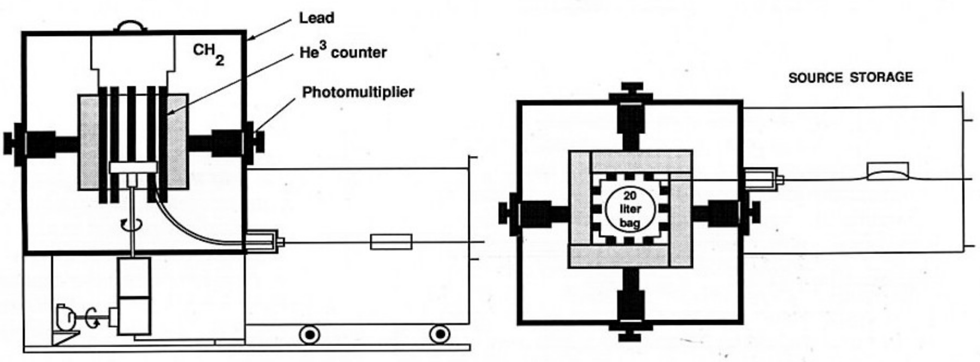

***Fig. 1.** Cosac system at CEN/Cadarache*

THE BANCO SYSTEM FOR 220-LITER DRUMS

The system is shown in a Fig. 2, which works in a mode of the passive analysis. The neutron counters and gamma-spectrometer simultaneously scan along a vertical axis of a rotated barrel. For mild matrixes with density 0,2 limits of detecting make about 10 mg of plutonium for fuel PWR and about 40 mg in a mode of coincidence. In active version the neutron-source from ^{252}Cf with intensity 10^6 n/s will be used. The neutrons of fission are detected ^{3}He-detectors. In this case limits of detecting make 30mg ^{235}U and 200 mg of plutonium for fuel PWR. A time of an exposition 1000 s make.

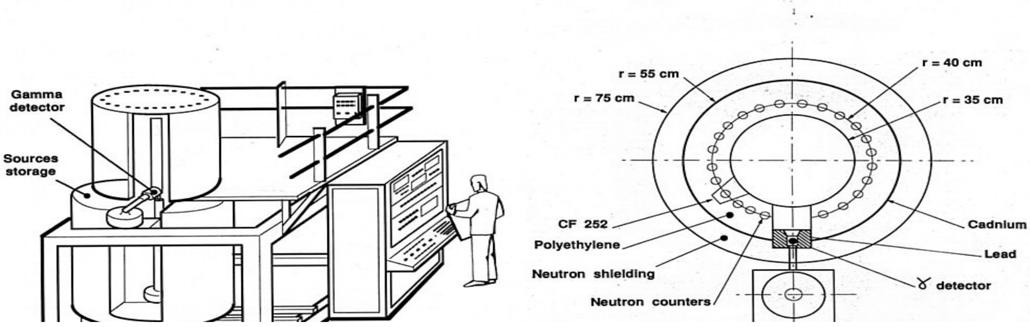

***Fig. 2.** Banco system at CEN/Cadarache*

5. NSC "KIPT"

In NSC KIPT on the basis of a linac complex of electrons LYE-300 for problem solving on detection of the contents of uranium isotopes and other fission stuffs in the RW the pulse neutron-source with intensity 10^{12} n/s was constructed. The trial experiments were conducted on the electron beam with energy E_e=20 MeV, mean current I_e = 2 μA and at frequency of 50 Hz. After a rejecting magnet the beam was routed on the leaden converter by thickness 4 cm, which served the neutron-making target [8]. After the target the moderator from polyethylene with the thickness of 8 cm was established. All-band detector Mc-Kibben was used for registration of neutrons of fission. Delaying neutrons of fission between impulses of the accelerator with time interval of 4 ms after a synchro-pulse were basically registered during this trial. As the RW was used 10 kg of sand with samples of uranium of different weight with 2% by enrichment. At an exposure time equal to 600 s the minimum contents equal to 0.012 g ^{235}U was registered which corresponds to concentration $1.2\cdot10^{-6}$ g/g.

The purpose of the following work was carrying out of experiments and development of techniques to analyze concrete samples on the contents of ^{238}U in them, which is necessary to solve problems linked with liquidation of Chernobyl catastrophe consequences [9,10]. The experiment was conducted under the same conditions, as the previous one. Concrete blocks with the weight of 0.5 kg with 2% enriched uranium introduced in them were used as samples. Samples from metal uranium 2% enriched were also used in the experiment as calibrating samples. From results of comparison of yields of the neutrons from concrete and calibration samples it was shown that under given conditions of the experiment it is possible to safely identify up to 10 g of ^{238}U in concrete. Now with the

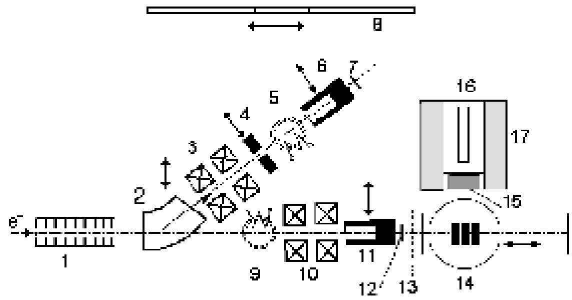

Fig. 3. *The scheme of the accelerator equipment*

1. Section of a linac.
2. Rejecting magnet (35°, removed).
3. Doublet of quadruple lenses.
4. Collimator - monochromator of a beam.
5,9. Inductive transient-time sensors of a current.
6,11. Faraday cups with removed absorbers.
7,12. Screens with fluorescing cover.
8. The trolley conveyor for moving of samples.
10. Doublet of short-focus quadruple lenses.
13. Ionization transient-time sensor of a current and position of a beam.
14. The removed neutron-making target air-cooled (target is established on mobile desktop).
15. Tested the sample of radioactive wastes.
16. Mc-Kibben detector.
17. Combined protection of the counter (paraffin, cadmium).

6. CONCLUSIONS

Now on the treatment to the RW the large development was received by industrial technologies, founded on application of neutron-sources on the basis ^{252}Cf and neutron generators. The electrons linacs will be used for the present only for debugging techniques on detection of small quantities ^{235}U and ^{239}Pu in different matrixes. Nevertheless, problem on definition of concentration is transuranic are rather varied both from the point of view of a radiation safety, and from the point of view of nuclear safety. The special value these problems have for Ukraine, as huge quantity of a firm and fluid radioactive waste of a nuclear industry and Chernobuly APS is accumulated, which require inventory. The pulse neutron-source with powerful neutron flux is indispensable for this purpose, which can be received on the electron linac. The experiments with large volumes of a radioactive waste (200 l) on beams of neutrons and gamma-quanta are now planned [11].

purpose of decreasing background the special stand on direct output linac (Fig. 3) is built. The experiments conducted on a hem with the same concrete samples, have allowed reducing detection limit ^{238}U up to 300 mg and on ^{235}U up to 1 mg which forms accordingly $6\cdot10^{-4}$ and $2\cdot10^{-6}$ g/g.

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