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TRANSLATION

SOME TECHNICAL ASPECTS OF THE NUCLEAR MATERIAL ACCOUNTING AND CONTROL AT THE NUCLEAR FUEL CYCLE FACILITIES

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ABSTRACT

The possibilities of nuclear material accounting and control are discussed at nuclear facilities of fuel cycle (WWER-type reactor, fuel fabrication plant, reprocessing plant and uranium enrichment facility) and zero energy fast reactor facility.

It is showed that for nuclear material control the main method is the accounting.

Possibilities and limitations of the application of destructive and non-destructive methods are discussed for nuclear

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Therefore, both the tested and considered methods suitable for measuring nuclear materials at the nuclear facilities material determinations at fuel facilities and their role in the accounting and safeguards systems as well as possibilities of the application of neutron method at a zero energy fast reactor facility.

INTRODUCTION

Power reactors of the WWER-type have been used extensively. At the present time they are being operated and constructed with the technical assistance of the Soviet Union in a number of non-nuclear weapon states.

According to the Non-Prolifiration of Nuclear Weapons Treaty, these reactor types are subjected to the safeguards system of IAEA.

In an effort to assist the Agency in applying the safeguards to the WWER-type reactor nuclear power plants the Soviet specialists have been carried out a number of investigations including the application of the accounting and experimental techniques for determining the actual amount of nuclear materials at Nuclear power plants as well as the studies of the inspection procedures at the plant, and under international transfer of nuclear materials [1-3]. In addition, certain technical problems such as the application of safeguards system to uranium enrichment facilities [4], utilization of probability methods for the safeguards purposes [5] have been also analyzed as well as many other studies concerning analytical provision of safeguards [6-7].

In order to further assist the Agency in developing safeguards technical problems, some technical aspects of accountancy and control of nuclear material at the fuel cycle facilities WWER reactor power plants

ЛИТЕРАТУРА

have been studied in the Soviet Union. The present paper describes the main results of these investigations.

1. ACCOUNTANCY AND CONTROL OF NUCLEAR MATERIAL AT THE WWER REACTOR FUEL CYCLE

The fuel cycle of WWER-type power reactors consists of a series of nuclear facilities such as a plant for fabrication of fuel rods and assemblies, reprocessing plant, and a uranium enrichment facility. The features of the fuel cycle essential to safeguards are as follows.

1. Low initial enrichment of fuel (3.6 - 4% mass), i.e. low strategical value,

2. High strategical value of the irradiated fuel due to plutonium production,

3. Using of nuclear materials both in bulk and cladding.

Table I shows an approximate distribution of the nuclear material over different nuclear facilities of the WWER-440 and WWER-1000 reactor fuel cycles[8] .

The above items of the WWER-reactor fuel cycle determine the system of control and safeguards applications scheme.

The accountancy system is based on the data obtained from weighing the transportation containers and analyzing the samples taken in the bulk form for nuclear material and calculation

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data (for reactors). This accountancy system makes it possible to present reports to the Agency about the acts of international transfers of nuclear material in the form of fuel assemblies from the Soviet Union to non-nuclear states and back to the USSR. The transportation is done by the V/O "Isotope".

Fig.1 shows a schematic drawing of safeguards applications at the WWER-reactor fuel cycle.

The nuclear materials in this fuel cycle are in claddings on the way from the storehouse of the fuel fabrication plant to the storage of the reprocessing plant, while then are in the bulk form in other instances.

The diversion nuclear material in the bulk form is possible either directly at the facilities or in the transfers between the corresponding MBA_s. In this part of the cycle, the main method of control is the accountancy for the nuclear material, which involves the weighing of the transportation containers and analyzing the nuclear material samples.

Containment and surveillance methods include sealing of the transportation containers, automatic photo-camera recording of the shipment and arrival of the transportation containers as well as the instruments making the recording.

Therefore, the errors in determining the actual amount of nuclear material in the bulk form can reach those required for the safeguards purposes.

The diversion of nuclear materials in claddings is possible only by replacing the entire fresh and irradiated assemblies by new assemblies since the fuel assemblies of the WWER reactor are designed so that they cannot be dasassambled and extraction of separate fuel rods cannot be done without destruc-

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22. Б.А.Бибичев, Л.И.Голубев, С.С.Коваленко, В.П.Майоров,

tion of the assembly.

In this part of the cycle, non-destructive control is the only method to measure the nuclear material in the assemblies, as a rule the error is now 3-5%.

Therefore, it seems expedient to establish the following additional requirements for this part of the cycle:

the assemblies should be identified by their serial numbers which remain unchanged from the moment of manufacture to destruction;

accounting for nuclear material and verification should be made separately for each assembly;

in addition to the information on uranium and plutonium contents in irradiated assembly the data on the average burn-up should contain.

For nuclear material in claddings the above requirements are sufficient to confine the control to the identification measurements.

The control of nuclear material over the eintire fuel cycle can be made in the following way:

verification of the correlation relations of the nuclear material fluxes moving between the corresponding material balance areas at the facilities;

calculation of the fluxes and distribution of the nuclear material over the fuel cycle, using the data on the actual reactor energy output,

The control and accounting for nuclear material described above are able to provide necessary accuracy in making balance of the WWER-reactor fuel cycle.

2 ACCOUNTANCY AND CONTROL OF NUCLEAR MATERIAL AT NUCLEAR FACILITIES

The actual amount of nuclear material in the fuel cycle is carried out in the material balance areas of nuclear facilities. In this connection, it is necessary, to consider the methods of performing such a control at the nuclear facilities of the WWER-reactor fuel cycle /at the nuclear power plant, fuel fabrication plant, fuel reprocessing plant, and at the uranium enrichment facility/.

Nuclear power plant. Nuclear material at the Nuclear power plant with the WWER-440 reactors is in the form of fuel assemblies. Non-irradiated fuel assemblies contain uranium dioxide with an initial enrichment up to 3.6% mass. Every irradiated assembly contains up to 0.8 kg of plutonium. Accounting for nuclear material is carried out both on the basis of the manufacturer nameplate and by calculational methods - on the basis of the actual reactor energy output [10].

The diversion of nuclear material can be possible either by replacement of fuel assemblies by new assemblies or irradiation of unacaunted fuel in the reactor, which escaped the Agency control.

Substitution of the assemblies can be made, for example, by showing the same assembly twice to the inspector, in different places of the Nuclear power plant (in the storehouse, in the reactor or storage). To prevent such diversions it is necessary to incorporate the following provisions:

three nuclear material balance areas corresponding to the storage of fuel assemblies should be formed;

MBA-1 MBA-2

Natural

identification of assemblies should be carried out by their serial numbers;

independent determination of the initial fuel enrichment should be made in non-irradiated assemblies and average fuel burn-up in irradiated assemblies. Possible locations of MBAs at a Nuclear Power Plant are shown in Fig.2c.

Calculation methods used for determining the quantity of nuclear material in assemblies are non sufficiently accurate. For example, the experimental analysis of the isotopic composition of the irradiated fuel in the assemblies of the WWER-365 reactor in the 2nd unit of Novo-Voronezh Nuclear power plant has shown that the deviation of the experimental values from the calculated values is ±1-2% /mass/ for uranium, ±5-6% /mass/ for plutonium, and ±2-3% for burn-up depths. The obtained results are compared in Figs.3 and 4.

Another way to control the burnup depth in the assemblies involves independent measurements of the reactor energy output. For this purpose a neutron flux integrator designed in the USSR can be used [11] .

To prevent diversional irradiation of unaccounted nuclear material in the reactor core it is necessary to seal the reactor cover for a period between the inspections.

In addition to these measures, automatic control of the process of arrival and dispatching the containers with fuel assemblies by photocameras should be established at the plant.

Fuel fabrication plant. At the fuel fabrication plant, nuclear material is either in the form of UO_2 powder with initial uranium-235 enrichment of 1.6±0.05% /mass/, 2.4±0.5%/mass/, and

3.6±0.05%, or as pellets with density of 10.2-10.7 g/cm^3, or as fuel assemblies with the uranium content of 120±1.5kg (for WWER-440) [12].

Nuclear material is accounted for by weighing the boxes containing UO_2 powder, pellets and fuel rods before and after they are filled with the material.

Diversion of nuclear material is possible during the movement of empty boxes as well as in transfers of the pellets for fuel rod filling (or disposing them as discarded or wastes).

To prevent such actions it is necessary:

to provide two nuclear material balance areas at the plant (MBA-I-fabrication of pellets; MBA-2 - fabrication of assemblies, Fig.2b);

to carry out the check weighing and analyzing of the samples of UO_2 powder and pellets;

photographic recording of the box receiving process and dispatching the containers with fuel assemblies;

sealing of the transportation containers.

Fuel reprocessing plant. Nuclear material at the fuel reprocessing plant is in the form of uranium and plutonium solutions [13]. The Uranium-235 content amounts to 10-11 kg/t of uranium, the plutonium content-8 kg/t of uranium for burnup 28500 MW/t of uranium.

Nuclear material is accounted for by weighing the received fuel, measuring the volumes and concentrations of the solutions.

Diversion of nuclear material can be made in removal of scrap, removal of uranium and plutonium solutions produced in technological reprocessing, and in waste solutions.

To prevent this it is necessary:

to provide four material balance areas: MBA-I - cutting and dissolving of the fuel; MBA-2 - separation of uranium an plutonium; MBA-3 - purification of uranium, and MBA-4 - purification of plutonium. (Fig.2d) ;

determination of the fuel mass before solving;

measuring the solution volumes and determination of uranium and plutonium concentrations in them;

determination of uranium and plutonium quantities lost with waste solutions.

The other methods for safeguards procedures are shown in Fig.I.

Uranium enrichment facility. Nuclear materials in the form of uranium hexafluoride of low initial enrichment are used at the uranium enrichment facility.The accountancy is based on weighing of the transportation containers and analysing the samples.

Diversion of nuclear material can made either by means of removal of its with empty cylinders or by enriching unaccounted nuclear material in the technological part of the facility.

For safeguards purposes it would be necessary that:

two nuclear material balance areas should be made (MBA-I- the storehause, and MBA-2 - the technological part, Fig.2a);

check weighing of the containers and analyzing the samples;

photographic recording of the arrival and dispatch of the containers;

Therefore, for safeguards purposes it is necessary to make the measurements of the following itemsat the fuel cycle facilities:

uranium dioxide powder;

uranium dioxide pellets;

fresh fuel assemblies;

irradiated assemblies;

uranium and plutonium solutions;

uranium-hexafluoride.

The techniques for measuring these materials are discussed below.

3. METHODS FOR MEASURING THE NUCLEAR MATERIALS.

Both non-destructive and destructive control of the above nuclear materials in the WWER-type reactor fuel cycle have been tested for the safeguards purposes.

Destructive analysis of the contents of uranium-235 and uranium dioxide include the gravimetric titrimetric, mass-spectrometeric, spectral photometry techniques [14-15]. The measurement results are presented in Table 2a. Variation coefficients of these methods were 0.1-0.3%.

The uranium content in the waste solutions was determined after purification by means of photometric or liminiscent methods with the variation coefficients of 2-3%.

After extraction the plutonium content was determined by the titrimetric or coulometric methods [16]. The variation coefficients in a 12-15 mg sample was 0.15%. These methods have been used in the PAFEX-I experiments [7].

The results are listed in Table 2b.

The plutonium content was determined also by the method of isotopic dilution with mass-spectrometric or alpha-spectrometric [17]. The variation coefficients of these methods were 0.2-0.3%.

TABLE II

a. ANALYSIS RESULTS OF THE URANIUM SOLUTIONS (% MASS)

Method	$UO_2(NO_3)_2$ solution		$UO_2(NO_3)_2$+Pu(%) solution	
	$\overline{X}$	$\overline{\sigma}$	$\overline{X}$	$\overline{\sigma}$
Potentiometric titration	100.0	0.05	99.93	0.08
Differential spectral photometry	100.3	0.20	100.10	0.30

б. ANALYSIS RESULTS OF THE PLUTONIUM SOLUTIONS (% MASS)

Method	PuO_2 powder		Plutonium nitrate solution	
	$\overline{X}$	$\overline{\sigma}$	$\overline{X}$	$\overline{\sigma}$
Coulometric	88.71	0.06	1.767	0.001
Radiometric	88.66	0.12	1.768	0.002

The plutonium content in the waste solutions was determined by dipping alpha-detectors into them. The variation coefficient was about 15-20% [18].

For safeguards purposes feasibility of the other methods described in Refs. [19-21] has been analysed.

To analyze the uranium content in scrap a modified Devise-Gray method with the variation coefficient 0.2-0.3% can be used. The same method can be also used in analysing the uranyl nitrate solutions obtained in the process of the irradiated fuel reprocessing of the WWER-reactors.

To analyze uranium hexafluoride the methods of gas mass-spectrometry or gravimetric technique can be used. In this case a sample of 80-120 g mass is taken, the variation coefficient is of 0.1%.

The mass of the fuel obtained for dissolving can be determined by weighing with account of undissolved material (0.02-0.03% mass). The calibration errors of the volume to be accounted for, measurements of the volume and sampling allow total uncertainty of about 1%.

Non-destructive control measurements of nuclear material have been used to determine the burnup depths in irradiated fuel rods and assemblies, the uranium-235 content in pellets and rods of the WWER-reactors.

The content of uranium-235 in non-irradiated pellets of enriched uranium dioxide was determined by gamma spectrometry. The measurement error was about 1%.

The same method with similar error was used to determine the relative uranium-235 content in non-irradiated fuel rods.

The burn-up and relative quantity of plutonium in irradiated assemblies were determined by gamma spectrometric measurements of the Cs^{134}, Cs^{137} content. The measurement error was about ±5% [22].

Fig.5 presents the measurement results made with an assembly of the WWER-365 reactor.

Neutron scanning of the assemblies is also an advanced method for such measurements [23].

The other methods of non-destructive control of nuclear material described in literature have been also considered [24-29]. The uranium-235 content in the transportation cylinders containing uranium hexafluoride can be also determined by using either the gamma spectrometry technique or measuring the number of neutrons from the F(n) reaction. The measurement errors can reach about 2-5%.

The gamma spectrometry technique can also be used to determine the uranium-235 content in boxes filled with enriched uranium dioxide powder. The error is 2-5%.

Determination of uranium and plutonium in solutions in measuring cells of reprocessing facility can be also promicing with an X-ray fluorescence method. The error to be expected should be 0.5±1.0%.

A pulse counting method of delayed neutrons from uranium-235 fission at bombarding the container with wastes by 14Mev neutrons can be applied to the wastes of the fuel reprocessing plant of the WWER-reactors. The variation coefficient is 15-30%. Plutonium content can be measured by recording of the delayed neutron coincidences from the spontaneous disintegration of Pu-240 with an error of 5-15%. For PuO_2 powder in the container, the Pu-240 spontaneous desintegration neutron coincidence method can be used with an error of 3-5%. The calorimeter method can be used with an error of about 1%. Non-destructive control methods have been also at the "fast" critical assembly "SPEKTR" of V.I.Lenin Research Institute, Dmitrovgrad [30]. Such highly enriched uranium (90% mass) or plutonium assemblies are not very "resistant" with respect to diversion, therefore they are of interest from the safeguards point of view.

The gamma-spectrometry methods have been found unsuitable for measuring the critical assembly packets because it is possible to imitate the nuclear material in the packets. The pulse-counting method of delayed neutrons in desintegration of uranium-235 bombarded by a neutron beam appeared more suitable. The error was ±2%.

To measure the actual quantity of uranium-235 in assembly an active neutron method was used. Constant decay measurements of the uranium-235 prompt neutrons are shown in Fig.6 and Table3.

Variations in α is 15-20% against the initial value while those of α from the point of uranium-235 extraction is 5%. The measurement error was about 1%.

TABLE 3.

CONSTANT DECAY VARIATION OF THE PROMPT NEUTRONS IN THE

Quantity and point of U-235 extraction (kg)	Weight of the displacing material (g)	Prompt neutron falling rate constant (sec^{-1})
Critical assembly with known quantity of uranium-235	-	4630 + 40
Critical assembly without 2.1 kg of uranium-235 extracted in the centre of the core(point A in Fig. 6)	1000g of polyethylene	3510 ± 25
The same at the periphery	800 g of polyethylene	3700 ± 20
Critical assembly without 1.5 kg of uranium-235 extracted (point B, Fig.6)	240 k of polyethylene and 2 steel rods	3930 ± 20

Therefore, both the tested and considered methods suitable for measuring nuclear materials at the nuclear facilities approximate in accuracy the safeguards requirements.

CONCLUSIONS

Studies of technical aspects of the accountancy and control of nuclear material at the fuel cycle nuclear facilities have shown that following:

the main method of safeguards is the accountancy for nuclear material at the fuel cycle nuclear facilities of the WWER-reactors. This method is based on the check measurements of the actual quantity of nuclear material in the material balance areas by weighing and analysing the samples;

analytical destructive control methods used for measuring the amount of nuclear material (titrimetric, coulometric, mass - and alpha-spectrometry methods and others) provide the accuracy of 0.5-1.0%;

non-destructive control methods used for measuring nuclear material (gamma-spectrometry, neutron measurements and others) provide the accuracy of 3-5% and require further improvoment.

As a whole, the nuclear material balance areas and methods described above for the control of nuclear material at the nuclear plant fuel cycle is able to provide the accuracy necessary for the safeguards purposes.

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Type of facility IAEA Safeguards	Nuclear power plant	Fuel fabrication plant	Fuel reprocessing plant	Uranium enrichment facility
Accounting method	1. Recording energy output of reactor	1. Control weighing of boxes containing UO_2 powder and analysis of samples 2. Control weighing and analysis of pellets	1. Control weighing of cut fuel 2. Control measurements of solution volumes and concentrations	1. Control weighing of transport cylinders containing UF_6 and analysis of samples
Storage method	1. Sealing reactor cover 2. Sealing the fuel-assembly containers	1. Sealing of boxes containing UO_2 and fuel-assembly containers	1. Sealing of containers with final U and Pu solutions	1. Sealing of transport cylinders
Surveillance method	1. Photographic recording of dispatch and arrival of fuel-assembly containers	1. Photographic recording of box-receiving and container-dispatching processes	1. Photographic recording of fuel-assembly containers 2. Photographic recording of dispatch of containers with solutions	1. Photographic recording of arrival and dispatch of transport cylinders

Fig.1. System of applying safeguards to nuclear facilities involved in the WWFR fuel cycle

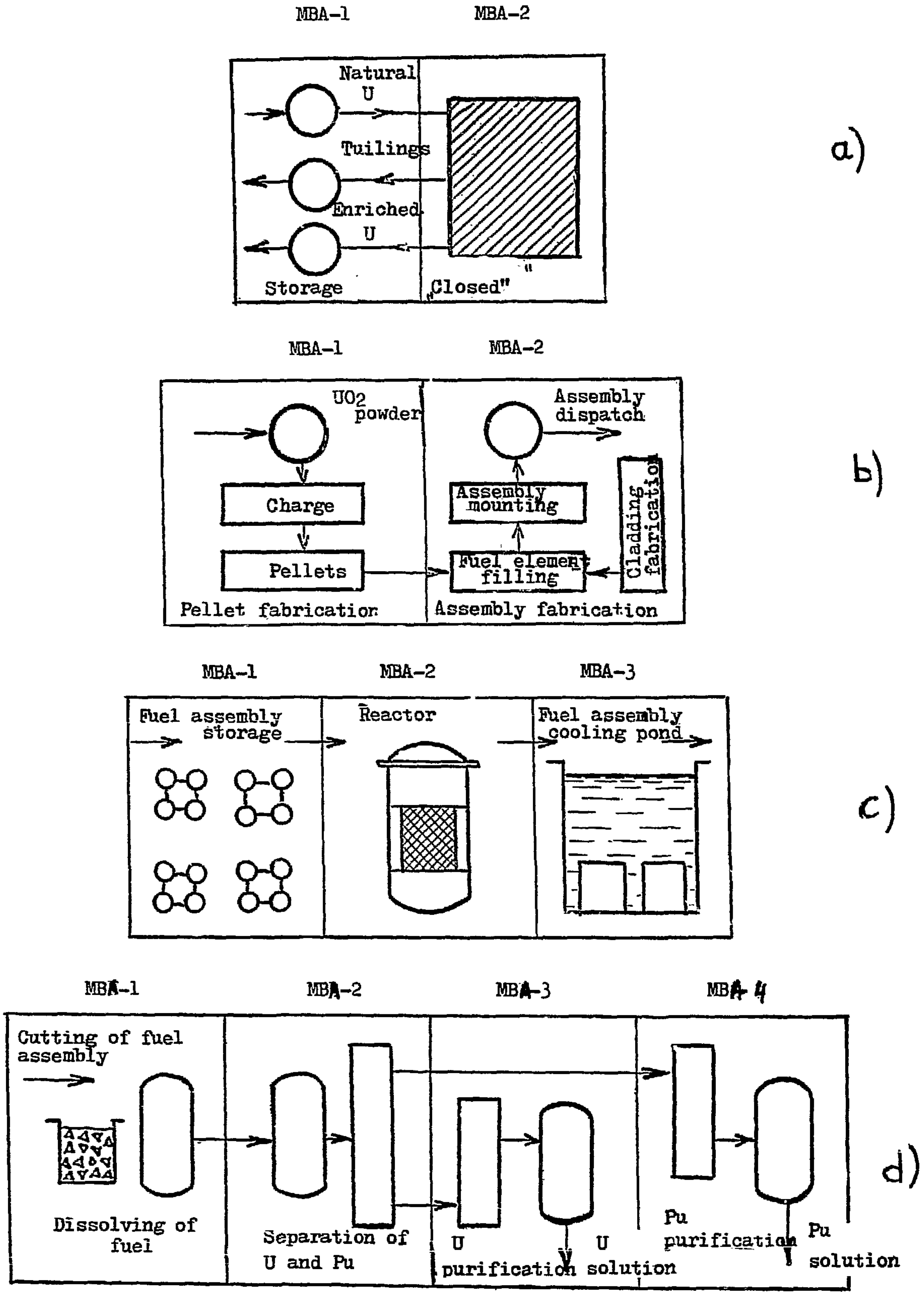

Fig. 2 Materials balance areas at facilities

a) Uranium enrichment facility
b) Fuel fabrication plant
c) Nuclear power plant
d) Fuel reprocessing plant

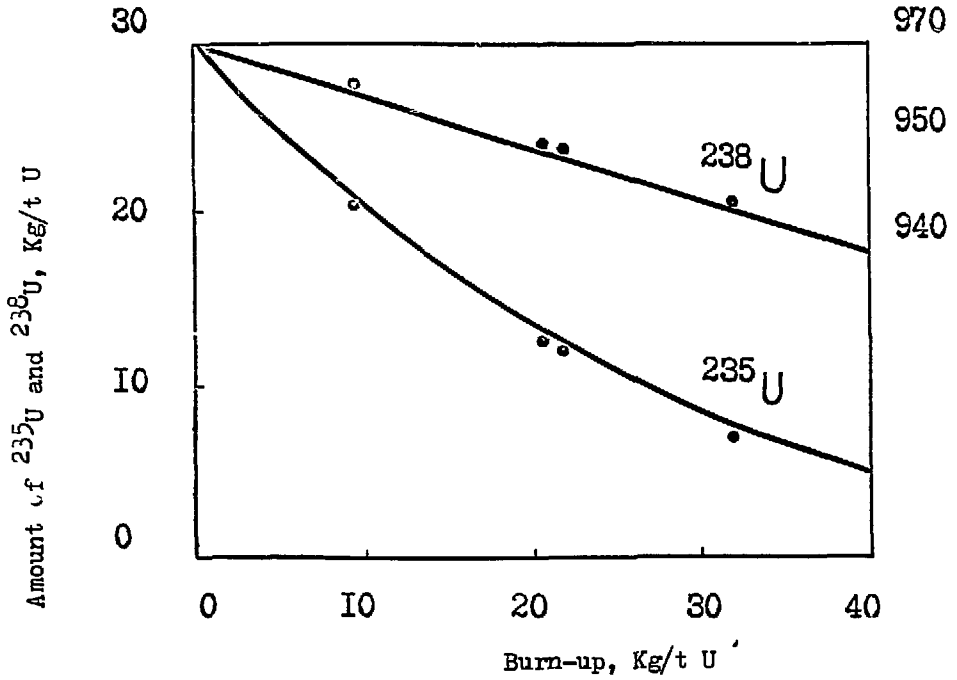

Fig. 3 Uranium content of WWER-365 fuel
(Initial enrichment - 3% mass)
—— - theoretical, ● - experimental

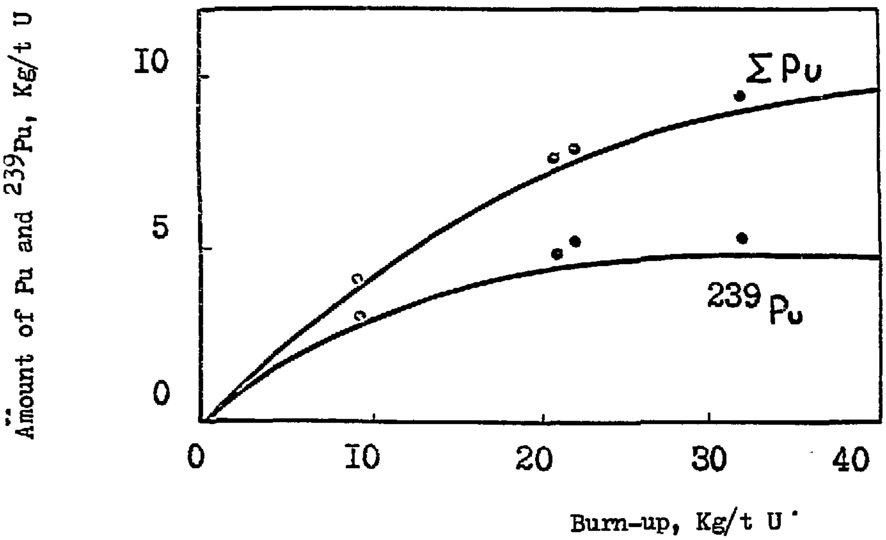

Fig. 4. Plutonium content of WWER-365 fuel
(Initial enrichment - 3% mass)
—— - theoretical, ● - Experimental

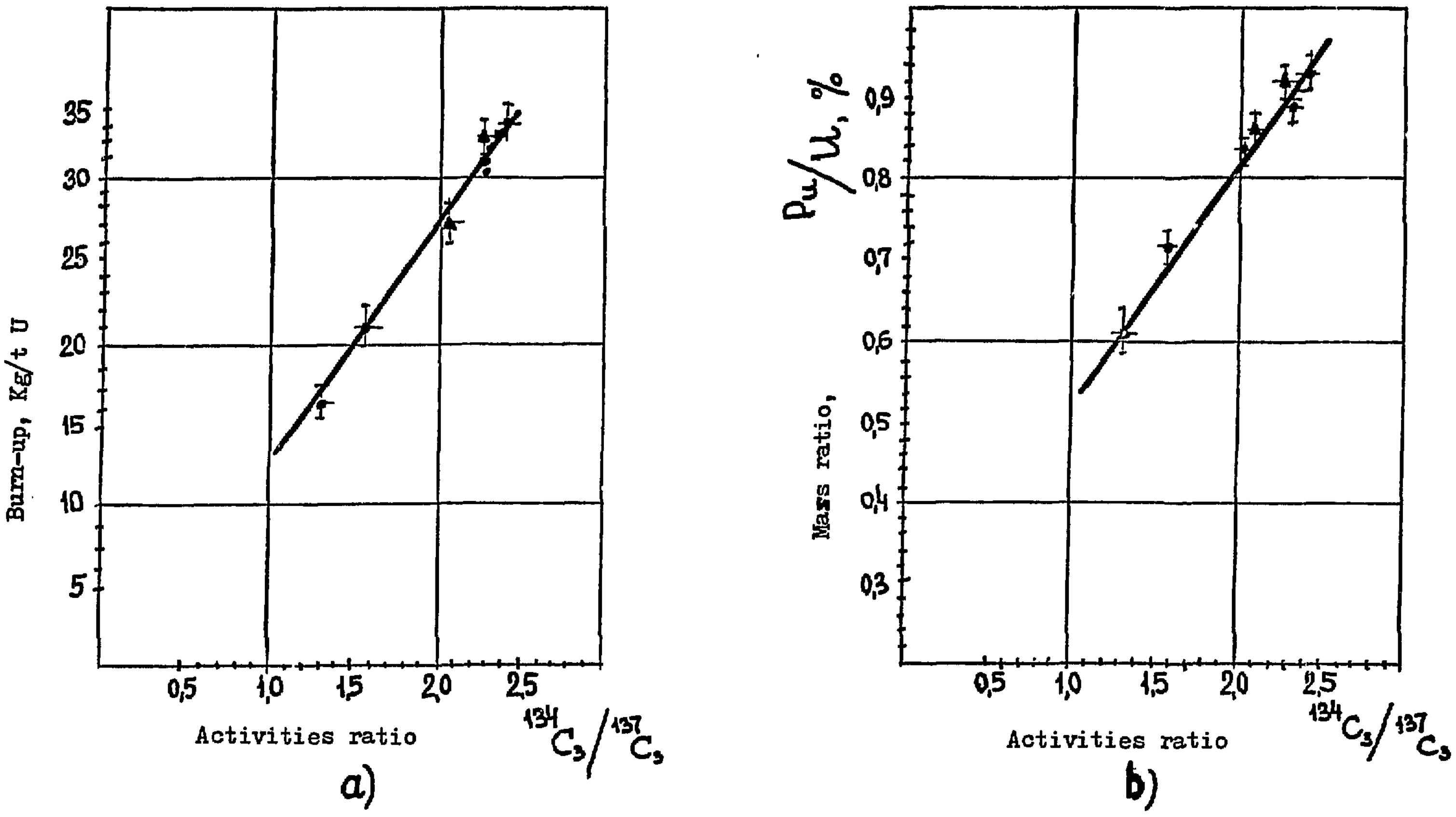

Fig. 5 Burn-up (a) and plutonium content (b) as a function of the ^{134}Cs/^{137} activities ratio for an irradiated fuel assembly of the WWER-365 reactor (Unit II of the Novo-Voronezh Nuclear Power Station)

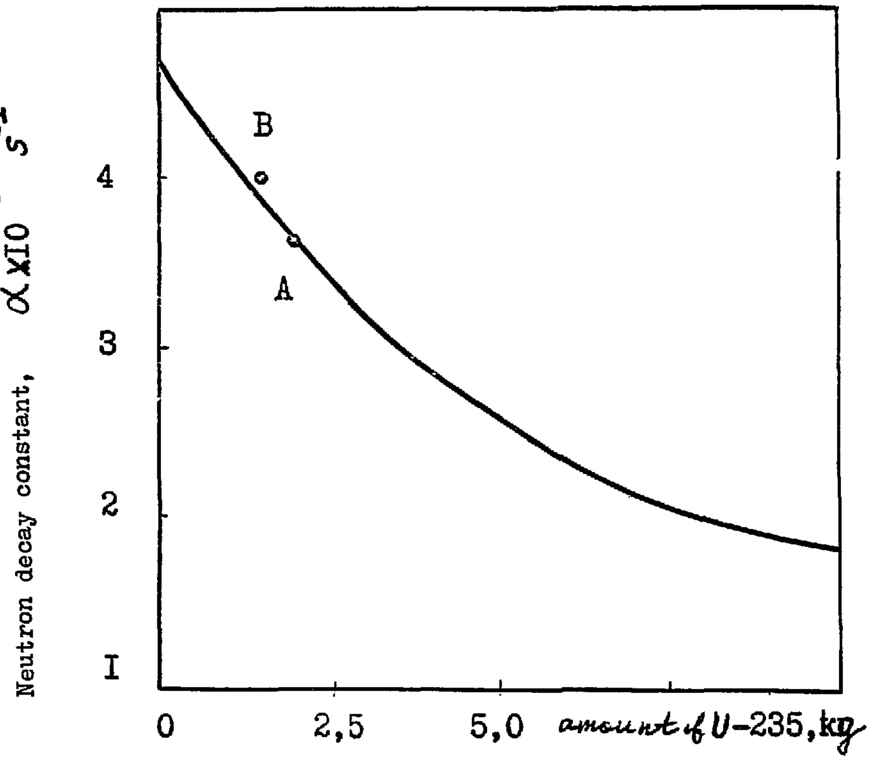

Fig. 6 Prompt neutron decay constant as a function of amount of extracted ^{235}U.