

# THE IRIS FACILITY AND NUCLEAR-SPECTROSCOPIC INVESTIGATIONS OF NUCLIDES FAR FROM THE BETA-STABILITY REGION

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## Introduction

The middle of 60s in different nuclear laboratories of the world was characterized by noticed growing of interest to nuclei lying far from the beta-stability line. Exotic properties and unusual physical phenomena were predicted by theoretical calculations for these nuclei. The extrapolation of nuclide mass values according to well-known semiempirical mass formulas led to the conclusion that the number of unknown nuclides was much more than that of known ones. There was a very promising situation to get much new information which could give the ground for new conclusions about nuclear structure and nuclear forces. The most interesting questions being discussed at that time were those about the possibility of discovery of proton radioactivity, delayed proton and neutron emission, delayed fission and other exotic decay modes which are energetically forbidden for nuclei close to the beta-stability line.

However, the effective production and investigation of nuclides far from the stability needed, because of their short lifetimes and low production cross sections, new experimental methods completely different from those used before. Several new ISOL (Isotope Separator On-Line) facilities, installed on beams of different type of bombarding particles (protons, neutrons and heavy ions), gave the possibility to get the mass-separated radioactive ions with very short (several milliseconds) lifetimes and with very low production cross sections.

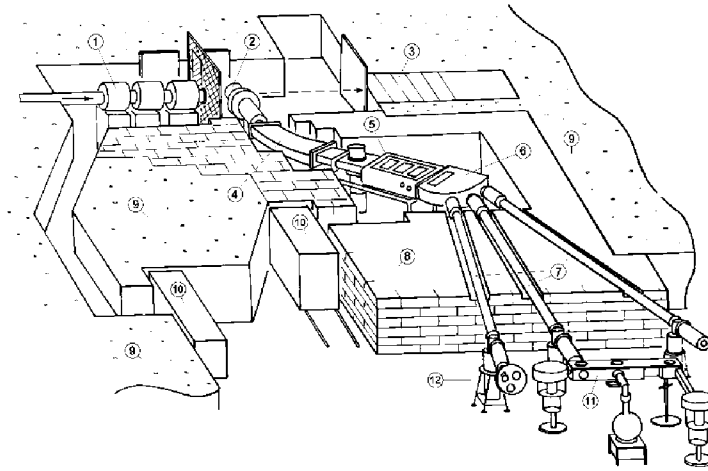
In Russia the first ISOL installation, named IRIS (Investigation of Radioactive Isotopes on Synchrocyclotron) [1], was built at Petersburg Nuclear Physics Institute (PNPI) in Gatchina under the leadership of Prof. E.Ye.Berlovich. For a long time it was the only working ISOL facility in Russia.

IRIS was put into operation in December 1975 on the proton beam of the PNPI synchrocyclotron, and first results were soon obtained [2]. For several following years, a large team of physicists from JINR (Dubna) took part in the investigations at the IRIS facility. The most important results of this collaboration were the determination of a large number of new masses of short-lived nuclei and identification of the proton drip line [3–8]. During the following years a number of new original ion source-target systems was developed, which allowed to produce and investigate both neutron-deficient and neutron-rich nuclei. Several experimental installations were put into operation, such as a laser nuclear facility, and a total absorption  $\gamma$ -ray spectrometer [9,10]. A high resolution mass-spectrometer [11] was designed and is being built at the IRIS experimental hall at the present time.

The Laboratory of short-lived nuclei, which uses the IRIS installation as its basic facility, collaborates successfully with well-known West nuclear centres, such as ISOLDE (CERN, Switzerland), GSI (Darmstadt, Germany), Universities of Marburg and Giessen (Germany), Cyclotron Laboratory of Jyvaskyla University (Finland), Rutherford Appleton Laboratory (England) and Neutron Laboratory in Studsvik (Sweden).

## The IRIS facility

The main part of IRIS is a mass-separator working on-line with 1 GeV proton beam of the PNPI synchrocyclotron. The general view of the system at the moment of starting its operation is shown in Fig. 1. The IRIS facility consists of the following parts: the proton beam line, the ion source-target unit, the mass-separator, radioactive ion beam lines and collectors, experimental installations equipped with different types of detectors, automatic-computer systems for accumulation and processing of experimental data.



**Fig. 1.** The general view of the IRIS facility. 1 – proton beam line; 2 – ion source-target unit; 3 – proton beam damper; 4 – mass-separator magnet; 5 – collector chamber; 6 – switch yard; 7 – ion beam lines; 8 – iron shielding wall; 9 – concrete shielding wall; 10 – iron gates; 11 – tape moving system; 12 – rotating collector.

### Proton beam line

The vacuum proton beam line includes 16 magnetic quadrupole lenses for proton beam tracing from the main accelerator hall to the target box of the IRIS building. The proton beam line is about 60 meters long. The proton beam intensity on the mass-separator target is about  $10^{12} \text{ s}^{-1}$ , the proton beam cross-section is  $1.5 \text{ cm}^2$ . The beam size is controlled with the help of proportional chambers installed along the beam line.

### Mass-separator

The sector magnet with the bending angle of  $55^\circ$  and the central ion trajectory of radius 1.5 m provides 10 mm dispersion for the ions with mass number 150. The acceleration voltage is 30 kV, the ion beam cross section in the focal plane is about  $1 \text{ mm}^2$ . There are three electrostatic deflectors in the switch yard, which bend ion beams by the angles  $30^\circ$ ,  $45^\circ$  and  $75^\circ$  into three ion beam lines, guiding ion beams to the IRIS experimental hall. For the purpose

of focusing and additional ion beam bending, the systems of triplet-quadrupole electrostatic lenses and bending deflectors are installed along every beam line. The mass-separator control desk includes the magnet power supply, the vacuum control system, the system of the ion source-target unit heating and the system of ion beam control and measurement.

### **Ion source-target unit**

The ion source-target unit is the main part of an ISOL system, therefore the development of high efficiency ion sources and targets was one of the most important tasks at the IRIS facility. The ideal target should fulfil many requirements. The most important of them are as follows:

1. High speed of reaction product release.
2. High thermostability.
3. Low pressure of the target material vapour.
4. Selectivity (possibility of the isobar separation).
5. Universality (possibility of production of a wide range of nuclides).

As some of these requirements contradict each other, it is clear that the ideal target development is a very difficult task.

The following types of targets were developed and used at the IRIS facility enabling to produce radioactive isotopes of the major part of the periodic table of elements:

a. A "boiling" target for the noble gas isotope production. It was used for the first IRIS experiments to produce xenon neutron-deficient isotopes.

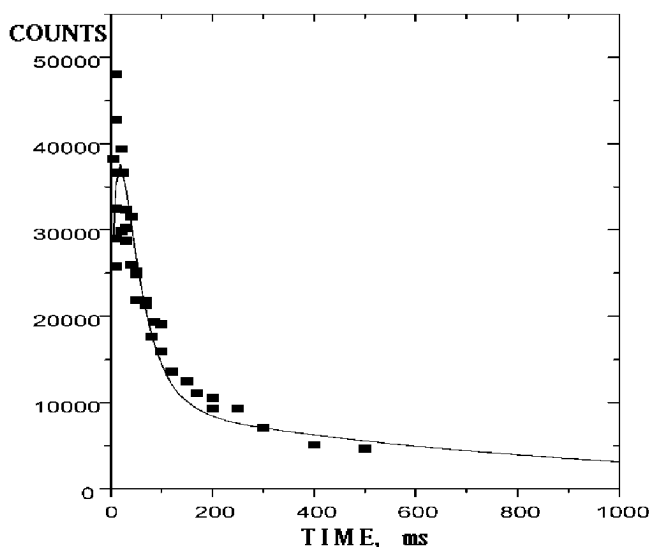
b. A molten lanthanum target for neutron-deficient Xe and Cs isotope production.

c. A tungsten or tantalum high temperature target heated by an electron bombardment. The target and the ion source are combined together in one unit. This target was worked out at JINR (Dubna) and was used in the experiments of IRIS (LNPI) – LNP (JINR) collaboration for neutron-deficient rare-earth isotope production. It was a very quick-acting target, but due to low value of the target material mass (2–3 g) the production yields were very low.

d. Refractory metal foil (Nb, Ta, W) targets for rare-earth, Rb, Sr, Y, J, and Cs isotope production [12]. These targets were developed in two modifications: the first one was the tantalum tape bundle which was welded at the ends of the target container in order to make the target ends hotter; the second one was made of rolled tantalum or tungsten foils of 20–30  $\mu\text{m}$  thickness enclosed in a tantalum container. The target mass was varied from 10 to 150 g. The working temperature was 2400–2600°C for a tantalum foil target and about 2200°C – for a niobium target. The shortest delay time of radioactive nuclides produced by this type of targets was about 1 s and the yields were up to  $10^7$ – $10^8$   $\text{s}^{-1}$ .

e. The target on the basis of pyrolysed diphtalocyanines of U, Th and other elements of III–IV group of the periodic table for production of both neutron-deficient and neutron-rich isotopes of a large group of elements [13]. Owing to highly porous, large surface of a carbon-like

material produced as a result of diphtalocyanine pyrolysis and also due to a high diffusion speed of atoms of some elements inside this compound, a fast record release was obtained for isotopes of some alkali elements. Such target was used at the Neutron Laboratory in Studsvik (Sweden) and at CERN (Switzerland) as well. In Fig. 2 the release curve of Rb from Th composed target is represented, which was measured during testing experiments at the ISOLDE mass-separator at CERN. The half-life time of Rb isotope release was about 30 ms.



**Fig. 2.** Time characteristics of ThCx target material .

f. The target on the basis of molten metal gold for production of refractory element isotopes of Hf–Pt group. Until now this group of elements is hardly available for production at ISOL facilities using massive targets. As it was shown by off-line experiments, while processing the surface of preliminary irradiated molten gold by some amount of SF<sub>6</sub>, the elements of Hf–Pt group are extracted completely from the gold target material as highly volatile compounds of fluorine [14]. However, this target was not used on-line due to its very low (lower than 0.1%) ionization efficiency.

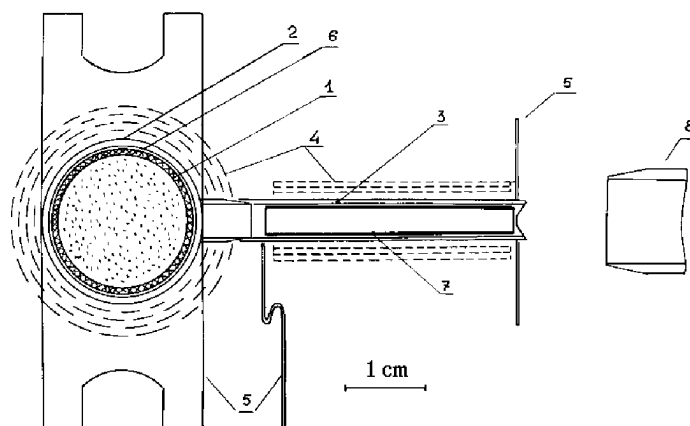
Four types of ion sources were used for on-line experiments at the IRIS mass-separator.

a. A standard Nielsen-type plasma ion source. It was used in the first on-line experiments for Xe radioactive atom ionization.

b. A high temperature ion source of surface ionization. It is the most widely on-line used type of an ion source at the IRIS facility. It can be efficiently used for the elements with ionization potentials up to 6.5 eV. For alkali metals (K, Rb, Cs, and Fr), the ionization efficiency is close to 100%.

c. A high selectivity and high efficiency ion source for negative ion production. It is a very high selectivity ion source which produces ions of halogens with 20–50% efficiency. Making use of this type of an ion source, iodine neutron-deficient and neutron rich isotopes were investigated.

d. A high efficiency laser ion source [15] with high selectivity ionization of different elements. This type of an ion source was developed for the first time and put into on-line operation at the IRIS facility. It can supply isobarically pure radioactive beams and, being used in combination with a mass-separator, gives possibility to get extremely purified ion beams needed for investigation of radioactive nuclides.



**Fig. 3.** Schematic drawing of a diphtalocyanine  $\text{MeC}_x$  ion source-target unit. 1 – target material; 2 – tantalum container; 3 – tungsten ion source; 4 – thermal screens; 5 – target and ion source connectors; 6 – tantalum foil ring; 7 – W–Re alloy foil for the ionization efficiency rise; 8 – extraction electrode.

### The spectrometric system and resources of computer-assisted analysis

Spectrometric devices of different type were used at IRIS including Si(Au) detectors for  $\alpha$ -particle measurements, Si(Li) detectors for  $\beta$ -particle and X-ray identification, Ge(Li) detectors for  $\gamma$ -radiation detection, Ge(HP) detectors made of high purity germanium for  $\beta$ -spectra measurements, and a large NaI  $\gamma$ -spectrometer with a cylindrical hole inside. Experiments with short-lived nuclei need many cycles of radioactive sources accumulation. Therefore, the amount of collected spectra can reach several thousands for one run of the experiment. The sorting of acquired information, the preliminary processing of experimental data and control of the experimental setup as well are being carried out during the course of the experiment.

## Nuclear-spectroscopic experiments with nuclides far from the beta-stability line

Detailed spectroscopic investigation of exotic regions of nuclides, making use of detectors of different type, allowed to identify new nuclides. The list of the new identified nuclei with some of their characteristics is given in Table 1.

Table 1  
Nuclides identified at the IRIS facility for the first time

Nuclide	Half-life, s	Identif. method	Nuclide	Half-life, s	Identif. method
$^{234}\text{Ra}$	30(10)	$\beta$	$^{159}\text{Lu}$	12.3(10)	$\alpha, \gamma, X$
$^{233}\text{Ra}$	30(5)	$\beta$	$^{158}\text{Lu}$	10(1)	$\alpha, \gamma, X$
$^{232}\text{Fr}$	5(1)	$\beta, \gamma, X$	$^{146}\text{Dy}$	32(5)	$\gamma, X$
$^{185}\text{Tl}$	19.5(5)	$\alpha, \beta, X$	$^{145}\text{Dy}$	18(3)	$\gamma, X$
$^{183}\text{Tl}$	9.7(6)	$\alpha, \beta, X$	$^{145}\text{Tb}$	29.5(15)	$\gamma, X$
$^{182}\text{Tl}$	2.8(6)	$\alpha, \beta$	$^{136}\text{Sm}$	42(4)	$\gamma, X$
$^{181}\text{Tl}$	3.4(6)	$\alpha, X$	$^{138m}\text{Pm}$	10(2)	$\gamma, X$
$^{163}\text{Lu}$	246(12)	$\gamma, X$	$^{131m}\text{Pr}$	6.5(10)	$\gamma, X$
$^{161}\text{Lu}$	72(6)	$\gamma, X$			

The set of criteria for new isotope identification is as follows:

- a. The nuclide mass number  $A$  is surely fixed due to the use of the mass-separator.
- b. The characteristic X-rays, arising in the electron capture or electron conversion processes, demonstrate the presence of the isotopes of the element being investigated.
- c. Simultaneous registration of amplitude spectra for different types of radiation ( $\alpha, \beta, \gamma$  and X-rays) allows to compare the obtained half-life values and gives additional confidence in the correctness of an unknown isotope and its radiation spectrum identification.
- d. The curve showing the time dependence of the intensity of the daughter nucleus radiation has characteristic "accumulation-decay" form; it testifies the existence of genetically connected unknown parent radiation in the spectrum. The use of this kind of curves gives the possibility to obtain the parent nucleus half-life.

### Nuclide mass determination by means of measurements of the positron spectrum endpoint energy

An unknown nuclide mass value can be determined if the mass of the daughter nucleus and the mass difference between parent and daughter nuclides are known. The mass differences between parent and daughter nuclei can be obtained by means of measurement of  $\beta^\pm$ -spectrum endpoint energies making use of a high-resolution beta-spectrometer.

Positron emitters in the region of rare-earth isotopes were studied at the IRIS facility. A high purity germanium crystal Ge(HP) with a volume 1.2 cm<sup>3</sup> was used as a detector.

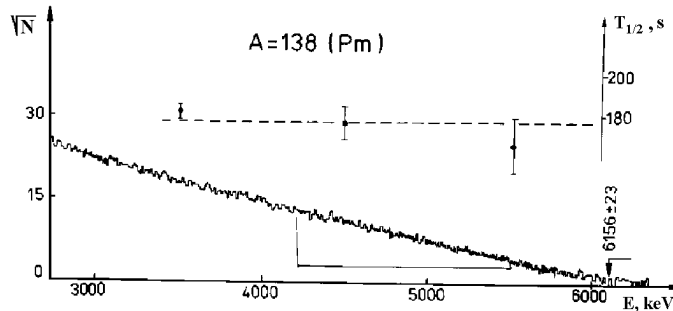
The value of resolution for 2 MeV electrons was 10 keV (the efficiency for high energy  $\gamma$ -ray registration by this  $\beta$ -detector was rather low).

The positron spectrum endpoint energies were obtained from Curie plot which was preliminary mathematically corrected taking into account spectrum distortions. For this purpose the nuclide positron spectra with well-known mass differences were measured in order to obtain the corrections to the spectrum endpoint energy determined as a result of the continuous  $\beta$ -spectrum processing.

The essential advantage of a Ge(HP) detector is the possibility of using it for the analysis of the composition of the measured source radiation due to simultaneous registration of  $\gamma$ -radiation with the energy  $< 1$  MeV and conversion electrons of a high multipolarity (or E0) transitions. Moreover, due to small dimensions of the detector it can be used in the experiment together with other detectors (Ge(Li) and Si(Li)) which also give information about the radiation composition and the background components.

The used methods of the measurement and processing of positron spectra allowed to perform for the first time with a good precision the determination of positron spectrum endpoint energies for a large range of masses. The complete set of data obtained is given in Ref. [7].

In Fig. 4, one of the measured positron spectrum of  $^{138}\text{Pm}$  is shown.



**Fig. 4.** The positron spectrum of  $^{138}\text{Pm}$  accumulated during one run of measurements. Marked by vertical lines is the region of energy which was used for the spectrum processing and for the endpoint energy extraction. At the top of the drawing the half-lives of different spectrum parts are represented.

### Identification of the drip line of the proton unstable nuclides

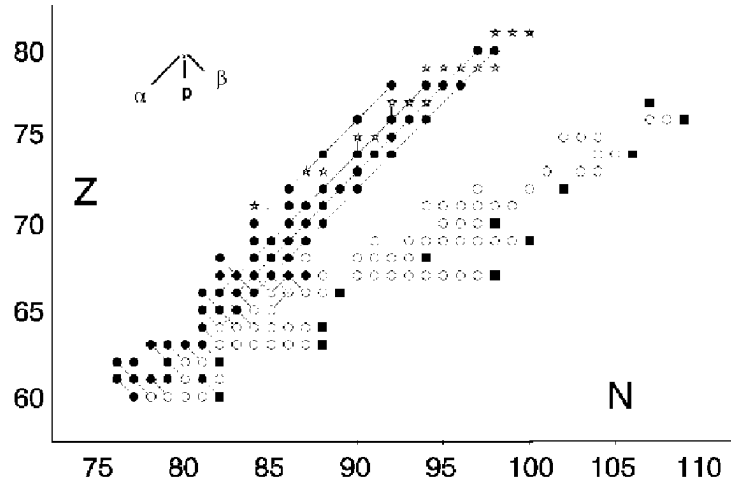
The nuclide mass determination, which was carried out using the measured endpoint energies of positron spectra, made it possible to construct the mass-surface of nuclides lying extremely far from the  $\beta$ -stability line. The precise measurements of  $\alpha$ -spectra in the rare-earth region [3,7] at the IRIS facility have led to observation of some unknown  $\alpha$ -emitters (Table 1). It allowed to close the long  $\alpha$ -decay chains and to determine all nuclide masses in these chains using the known mass values of nuclides lying at the end of every chain.

The mass values obtained made it possible to identify the proton-unstable nuclei making use of the relation:

$$S_p = M(Z - 1) + m_H - M(Z, N),$$

where  $S_p$  – the valent proton separation energy,  $M$  – the nuclide mass.

Nuclei for which the energy turned out to be negative are shown in Fig. 5 by asterisks.



**Fig. 5.** The fragment of the nuclide chart with a region of nuclides whose masses were determined for the first time at IRIS. Solid circles are nuclides with masses measured at IRIS, open circles are nuclides with known masses, squares represent stable nuclides. The proton-unstable nuclei are marked by asterisks.

We consider it to be the first experimental identification of the drip line of the proton unstable nuclei.

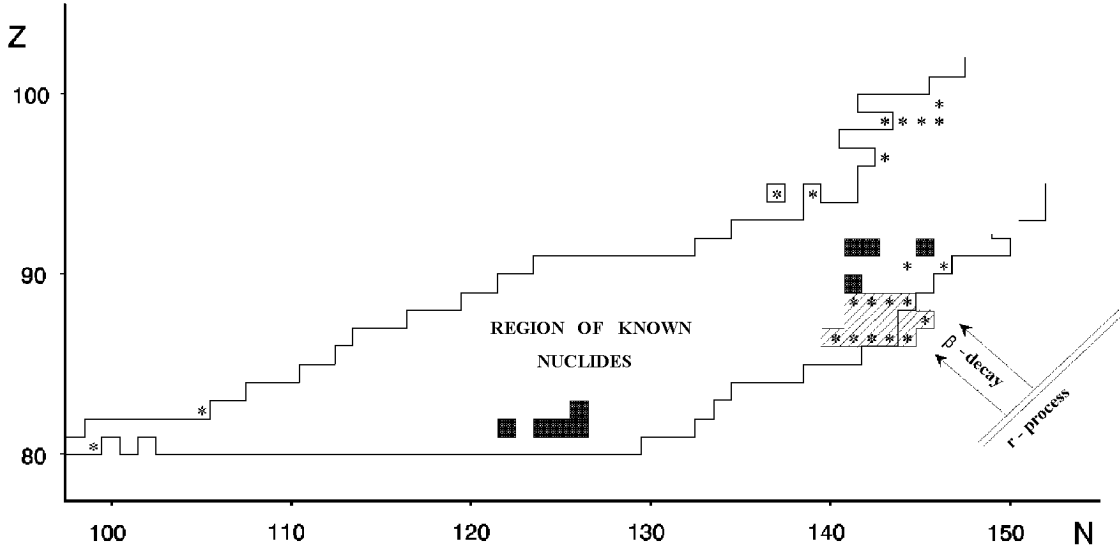
### Search for delayed fission of neutron-rich nucleus and the cosmochronology

As it was pointed out [16], there was a large region of heavy nuclei where a new type of decay – a delayed fission (fission after  $\beta$ -decay) could be found. There was a large amount of theoretical works since that time, which gave the probability of this kind of decay in different nuclides. Rather optimistic estimations have been made in one of the latest articles of H.V.Klapdor (1985) in respect to the position of the delayed fission boundary line on the nuclide chart.

In accordance with these calculations, for nuclide  $^{232}\text{Fr}$ , which is the closest to the region of known nuclei, the branching ratio of the delayed fission is 29%. Taking into account this value, which considerably changes the population balance of the cosmochronometer  $^{232}\text{Th}$  in the  $\beta$ -decay stage of the nuclear synthesis in nature, the estimation of the age of the Universe was carried out by H.V.Klapdor [17]. It turned out to be  $T_U = 22^{+2}_{-5} \times 10^9$  years, that is higher than the estimation given earlier  $T_U \leq 15 \times 10^9$  years.

At the IRIS facility the search for the delayed fission of  $^{232}\text{Fr}$  and neighbouring nuclides was carried out [18]. Until now it has been the only attempt. The search for the delayed fission

was carried out in parallel with the identification of  $^{232}\text{Fr}$ ,  $^{233}\text{Ra}$  and  $^{234}\text{Ra}$  isotopes which were not known before. For the registration of fission fragments the surface-barrier Si(Au) detector was used. The search region for the delayed fission is shown in Fig. 6, the branching ratios are given in Table 2.



**Fig. 6.** The nuclide chart fragment. Marked by asterisks are nuclei for which the branching ratios of the delayed fission were measured. Solid squares represent stable and long-lived nuclei and nuclides-cosmochronometers as well. Shaded area shows the region of the search for delayed fission at IRIS.

Table 2  
Spectroscopic information about nuclides from the cosmochronological region

Nuclide	$T_{1/2}$ , s	$Q$ , MeV	Radiation	Intensity, %	$P(df)$
$^{226}\text{Fr}$	48	3.7(1)	$\beta, \gamma$	-	-
$^{228}\text{Fr}$	39	4.2	$\gamma 474$	20	$< 10^{-7}$
$^{230}\text{Fr}$	19	-	$\gamma 129$	100	$< 10^{-6}$
$^{232}\text{Fr}$	5(1)	-	$\gamma, \beta, X$	100	$< 10^{-6}$
$^{232}\text{Ac}$	35	3.4(1)	$\gamma 665, X$	15	$< 10^{-6}$
$^{233}\text{Ra}$	30(5)	-	$\beta$	100	-
$^{234}\text{Ra}$	30(10)	-	$\beta$	100	-
$^{234}\text{Ac}$	44	4.6(15)	$\gamma 688 + 693$	100	$< 10^{-4}$

As one can see from Table 2, there is no any mentioned above cases where the fission fragments were found. For nuclide  $^{232}\text{Fr}$  the limit on the delayed fission branching ratio is  $\leq 2 \times 10^{-6}$ . It is considerably lower of the calculated estimation 0.29 and shows that the delayed fission of  $^{232}\text{Fr}$  could not block the formation of cosmochronometer  $^{232}\text{Th}$  during  $r$ -process and, hence, the theoretical estimation of the age of the Universe made by H.V.Klapdor has to be revised.

Spectroscopic investigations carried out at the IRIS facility during 20 years period are more numerous than cited above. We did not mention, for example, the works devoted to the behaviour of proton and neutron pairing energy which has a tendency to simultaneous rise when approaching to the proton drip line.

Several works were devoted also to renormalization of the constant of the weak axial-vector current  $G_A$  in nuclei. The experimental information about single Gamov-Teller  $\beta$ -transitions in the "magic" nucleus  $^{146}\text{Gd}$  region allowed to obtain experimental values of the partial transitions probabilities and, making use of the "magic" nature of nucleus  $^{146}\text{Gd}$ , to calculate these probabilities. From the comparison of experimental and theoretical values the estimation of effective  $G_A$  constant was made. Comparing the renormalized  $G_A$  constant for nuclei and the effective constant of pion-nucleus interaction  $G_{\pi A}$ , which were derived from two completely different processes, one may conclude that the requirements of the partially conserved axial current (PCAC) theory are fulfilled in nuclear matter [19].

Out of this review are the investigations of the beta strength functions carried out at IRIS with the help of the total absorption gamma spectrometer (see, for example, Ref. [20]).

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