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Review Article

State-of-the-art progress of gaseous radiochemical method for detecting of ionizing radiation

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ABSTRACT

The article provides a review of the research results obtained during of more than 20 years concerning using the gaseous radiochemical method (GRCM) for detecting of ionizing radiation. This method based on threshold nuclear reactions with production of radioactive noble gas which does not interact with the materials of gaseous tract. The applications of GRCM in the diagnostics of neutrinos, neutrons, charged particles, thermonuclear plasma thermometry, and the study of the structure and dynamics of astrophysical objects, position-sensitive dosimetry of neutron targets with accelerator driving, spatial distribution of the fast neutron flux density in a nuclear reactor allowing the transformation of longitudinal coordinate of neutron flux distribution into a temporal distribution of the radiochemical gas decay counting rate ("barcode" semblance) and measurement of bombarding particles spectra are described. Experimental testing of the described technologies was made on the neutron target driven with the linear proton accelerator of Institute for Nuclear Research of Russian Academy of Sciences (INR RAS).

1. Introduction

The main methods for detecting of ionizing radiation uses detectors in which the decay counting volume located in close proximity to the area of interaction of incident particles with the target material. The GRCM is characterized by the presence of an intermediate agent, a carrier gas, transporting radioactive gas - product of a nuclear reaction from the interaction region to the decay counter. This separation can significantly reduce the background signal in the counter. The use of noble gases both as a gas-carrier and radioactive product decaying in the counter allows on the one hand avoiding the interaction of these gases with the structural materials of the gaseous pipeline and on the other hand to transfer the decay signal in remote counter. To register ionizing radiation in real time (monitoring), a flowing gas radiochemical method was proposed [1–4]. A similar method was previously used to detect the decays involving neutrinos in a chlorine – argon detector [5], and also to measure the background of fast neutrons with very low fluxes of ~10–2 cm-2day-1 in a gallium – germanium detector [6,7]. A feature of these measurements was the exposure of large (100–1000 kg) masses of the liquid active substance tetrachlorethylene C2Cl4 under neutrino and neutron fluxes, followed by condensation of the accumulated radioactive noble gas at low temperatures. The installation scheme for extracting of radioactive argon from the detector is shown in Fig. 1.

Helium is pumped through a barrel of tetrachlorethylene, which entrains the radioactive noble gas 37Ar, which was formed as a result of the nuclear reaction 37Cl (νe, e−)37Ar. At the same time, a mixture of gas -carrier He with 37Ar is accumulated in the upper part of the barrel. The gaseous mixture then passes through a refrigerator at a temperature −40 °C. In this case, most of the C2Cl4 vapor condenses. Then the gas passes through a microfilter trap, where the remaining vapor of C2Cl4 settles. Dry gas passes through a heat exchanger into which an absorbent carbon trap is placed. The trap is cooled by means of liquid nitrogen so argon and some other gases are adsorbed inside, but helium freely passes on and gets back into the barrel. When almost all argon from the detector is trapped with activated carbon, the trap is heated up to ~200 °C, gas is pumped out of trap and goes to a quartz furnace with titanium powder heated up to 900 °C. Under these conditions, active impurity gases namely O2 and N2 form titanium oxides and nitrides correspondingly. The remaining gas is a mixture of noble gases. Argon is extracted from this mixture by the method of gaseous chromatography. To determine the amount of produced gas, the condensate was placed in a proportional stationary counter. A similar method was used [8] when measuring the fast neutrons

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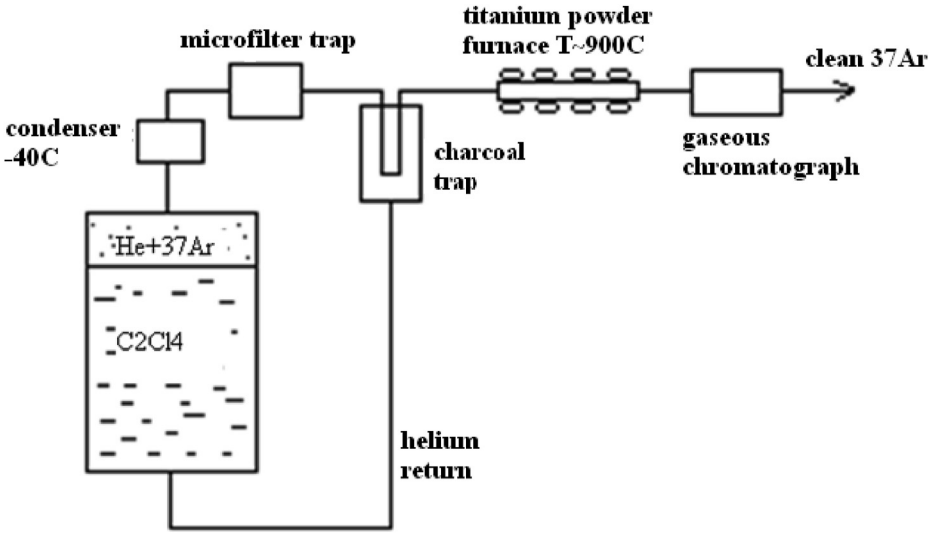

Fig. 1. Installation scheme for the extraction of noble radioactive argon produced in nuclear reactions.

background in the GALEX experiment [9,10].

A feature of the flowing gas radiochemical method is the use of the property of free output of noble radioactive gases from the crystal lattice of some solids. The installation scheme of a GRCM with a solid-state active substance is presented in Fig. 2. The noble radioactive gas formed in the detector ampoule is transported by the gas - carrier into a proportional flow-type gas counter, where the decay rate of the nuclei of the noble radioactive gas is measured, which is uniquely related to the ionizing radiation flux density in the detector ampoule. The registration of ionizing radiation fluxes is carried out by a proportional flow counter, which allows real-time measurements. Note there is the time delay between gas production and its registration caused by the time of gas transfer in the pipeline.

In the stationary regime of irradiation of the ampoule (with a constant flux of ionizing radiation Φ and a constant gas flow L of the carrier gas), all newly formed activity above the equilibrium is removed from the volume of the ampoule by the flow of the gas-carrier. Therefore, the specific activity of the gas - carrier is related with the flux of ionizing radiation by the ratio:

$$I = \frac{V_c P_c N 0 \sigma \Phi}{1.44 t_{1/2} L}\bullet, \tag{1}$$

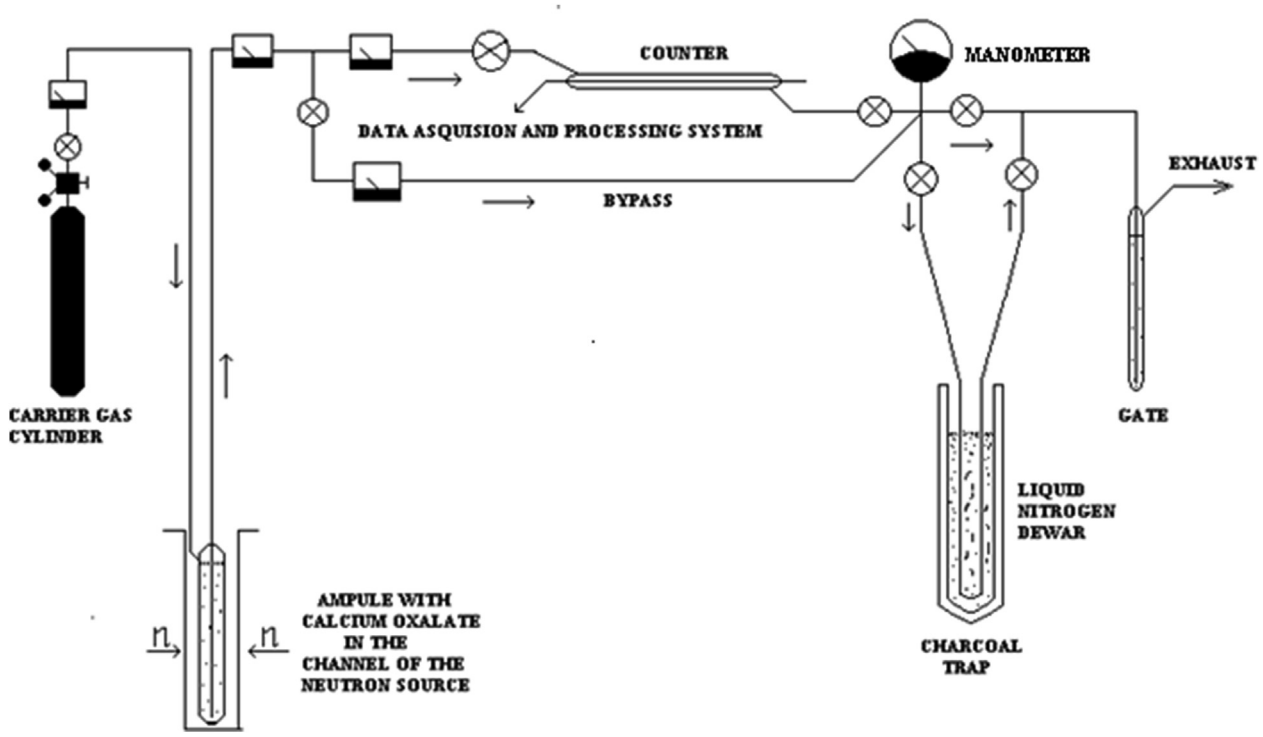

Fig. 2. Installation scheme of a radiochemical detector with solid-state active substance.

where I is the number of decays per second of radioactive inert gas in the counter, Vc is the working volume of the counter in cm3, Pc is the pressure in the counter in physical atmospheres, N0 is the number of atoms of the active substance in the ampoule, t1/2 is the decay half-life of the noble radioactive gas in seconds, σ is the cross section of the nuclear reaction of production of a noble radioactive gas averaged over the spectrum of ionizing radiation, Φ is the flux of ionizing radiation (cm-2s-1), L is the gas flow in cm3s-1. Eq. (1) is applicable for Va/L ≪ t1/2, where Va - is the volume of the ampoule with the active substance. As can be seen from Eq. (1), the proposed method for detecting of ionizing radiation is absolute and does not require calibration, since all quantities in Eq. (1) can be measured with high accuracy. In addition, one can list other advantages of this method:

- insensitivities to γ - quanta (radioactive gas decay counter can be removed far enough and shielded);
- relatively fast response time (few minutes) to a change in the flux of ionizing radiation (determined by the gas exchange rate in the ampoule, the diffusion coefficient of the radioactive gas - product in the crystallite of the active substance, and the length of the transport path);
- lack of moving parts, and, as a result, simplicity and reliability in operation. Unlike any other circulation scheme in which the active substance moves along the circuit (and, accordingly, the total activity is equal to the integral over the circuit), the production rate of radioactive inert gas is determined by the ionizing radiation flux density at the location of the ampoule with the active substance;
- in the case of a solid state of the active substance its vapor pressure is low, then there is no transfer of the active substance by transport gas;
- the absence of a liquid phase automatically solves the problem of a low gas exchange rate (at sparging the liquid) between the carrier gas and the active substance.

GRCM was used to monitor the neutron flux of a RADEX pulsed neutron target [11–13] with driving from a linear proton accelerator of the Moscow meson factory of the Institute for Nuclear Research of the Russian Academy of Sciences (INR RAS). A sketch of the RADEX facility in a section view is presented in Fig. 3. The proton beam from the ion guide passes through the aluminum first wall of the cylindrical ampoule and enters the active zone of the target, assembled from titanium-coated tungsten plates, cooled by light water. Inside the core, at a depth of ~4 m from the upper flange and at a distance of ~40 mm from the first wall, there is a cylindrical irradiation channel with a diameter of 73 mm. As can be seen from Fig. 3, an ampoule with an active substance of calcium oxalate of natural composition was placed in the irradiation channel of the RADEX installation.

During the time elapsed after the first experiments, the work was carried out to study possible applications of GRCM in various fields of fundamental and applied science in such areas as:

- neutron diagnostics
- diagnostics of charged particles
- diagnostics and thermometry of thermonuclear plasma,
- study of the structure and dynamics of astrophysical objects,
- position-sensitive dosimetry,
- measurement of the spectra of bombarding particles.

1.1. Flux monitoring and spectra measuring of bombarding particles

To monitor the fast neutron fluxes at the RADEX facility, reaction 10 (with calcium oxalate) from Table 1 was used. An illustration of neutron flux monitoring with GRCM is shown in Fig. 4. Here, the dependence of the decay count vs. time is presented over an interval of three days of continuous registration in automatic mode. Dark areas are colored corresponding to the presence of a proton beam on the target.

Neutron spectrometry with the help of GRCM based on various nuclear reaction thresholds can be carried out using several (ideally as many as possible) reactions from Table 1. It is interesting to note that 5 reactions namely 9, 10, 11, 12, 13 and 14 with different thresholds are possible on calcium natural mixture. The simplest fast neutron spectrometry can be carried out already on one active substance - calcium oxalate. In experiments on the RADEX pulse target, two of the indicated reactions, namely 9 and 11, were recorded. Fig. 5 shows the decay spectrum in the proportional counter obtained as a result of measurements pointing above. The k-peak of 37Ar decays in the 280 channel is clearly seen, corresponding to the energy of 2.9 keV. The low-energy peak is due to beta decay of 41Ar. It can be seen from Fig. 5 that the spectrum from beta decay decreases smoothly with increasing energy; therefore, the contributions from 37Ar and 41Ar can be effectively separated, demonstrating an example of the simplest fast neutron spectrometry at the RADEX facility.

In addition to neutrons, the GRCM allows the detection of protons, deuterons, tritons, and possibly heavier nuclei if their energies are high enough allowing the nuclear reactions producing the radioactive noble gases. Table 2 shows, as an example, nuclear reactions involving protons producing inert radioactive gases.

As can be seen from Table 2 there is the principal possibility of proton spectrometry in the energy range of threshold nuclear reactions between 1.5 and 55 MeV. For the reactions 1 and 2 from Table 2, suitable chemical forms of the active substance are known: fluorohexane C6H13F and chlorhexane C6H13Cl, respectively. The radioactive noble gases 19Ne and 37Ar formed in the liquid medium are extracted according to the scheme shown in Fig. 1 in which fluorohexane or chlorhexane are used instead of tetrachlorethylene.

2. Diagnostics of thermonuclear plasma

From the point of view of the possibilities of the GRCM described above, monitoring of 14 - MeV neutrons in thermonuclear installations is possible using practically any of the reactions listed in Table 1. However, this method cannot be used in cases where the monitor should give a control action for a time shorter than the minimum signal delay time in a GRCM. A change in plasma temperature in a fusion reactor causes an increase in ion energy due to the Doppler effect. It was shown in Ref. [14] that in the case of the Maxwell plasma, the neutron spectrum near 14 MeV has a Gaussian shape. In this case, the width of the energy distribution of the neutron pulse $\Delta En$is related to the plasma temperature Tp by the relation [15]:

$$\Delta En = 178\sqrt{Tp}, \quad (2)$$

where $\Delta En$and Tp are expressed in keV. The cross section of a nuclear reaction, the result of which is the production of a noble radioactive gas, depends on the energy of the incident neutrons. Earlier, a neutron gaseous detector based on the Bragg effect was proposed for thermonuclear plasma thermometry [16]. The operation principle of this detector is based on registration of decays of fusion products - alpha - particles and neutrons produced in the synthesis reaction $2H + 3H \rightarrow 4He + n + 17.6$ MeV. These products ionize the gas mixture and produce the tracks in a proportional counter and stop due to the Bragg effect. Among these tracks

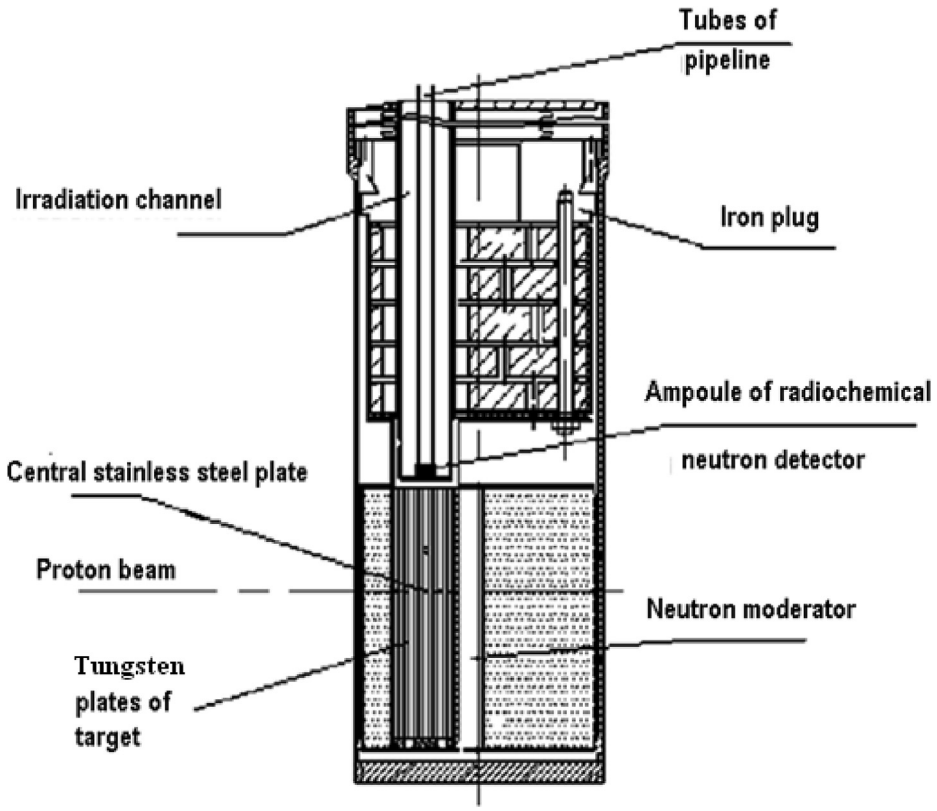

Fig. 3. Scheme of the RADEX facility in the experimental complex of the INR RAS with an installed gaseous radiochemical neutron detector.

Table 1
Characteristics of nuclear reactions involving neutrons, the products of which are noble radioactive gases.

№	NUCLEAR REACTION	ISOTOPE CONTENT	HALF LIFE OF GAS PRODUCT	REACTION THRESHOLD, MeV	CROSSSECTION AT 14.5 MEV MILLIBARN
1	6Li(n,p)6He	7.5%	0.8 s.	3.2	5.8
2	7Li(n,np)6He	92.5%	0.8 s.	11.4	10
3	7Li(n,d)6He	92.5%	0.8 s.	9.0	11
4	9Be(nα)6He	100%	0.8 s.	0.7	10
5	23Na(n,p)23Ne	100%	37 s.	3.8	30
6	26 Mg(nα)23Ne	11.1%	37 s.	7.0	85
7	39K(n,p)39Ar	93.3%	269 years	1.0	385
8	41K(n,p)41Ar	6.7%	1.83 h	3.0	45
9	40Ca(nα)37Ar	96.94%	35 days	1.0	130
10	42Ca(nα)39Ar	0.6465%	269 years	2.5	84
11	44Ca(nα)41Ar	2.09%	1.83 h	6.0	23
12	46Ca(nα)43Ar	0.0035%	5.37 min.	6.0	2
13	48Ca(nα)45Ar	0.19%	21.5 s.	14.0	0.5
14	85Rb(n,p)85Kr	72.17%	10 years	2.5	48
15	87Rb(n,p)87Kr	27.83%	76 min.	3.1	13
16	88Sr(nα)85Kr	82.6%	10 years	9.0	3
17	133Cs(n,p)133Xe	100%	5.25 days	6.0	19
18	138Ba(nα)135Xe	71.7%	9 h	7.5	2.5

selection has been made such that tracks completely located inside the counter volume. Such selection was made to exclude cases of tracks passing through the counter's walls. Plasma thermal heating during the synthesis reaction causes a broadening of the neutron spectrum line, which is directly determined by analyzing the current signals of the proportional counter. The disadvantage of this detection method is its high background load due to location of counter near the active zone of the thermonuclear reactor. The GRCM is practically free from the influence of background events due to its "sharpening" on a well-defined nuclear reaction and a specific product - a noble radioactive gas. In addition, in the case of GRCM detector, the counter can be removed far from the active zone and equipped with shielding against background radiation.

The Doppler shift to the energy Eq. (2) changes the cross section of the nuclear reaction. If such a change in the magnitude of the cross section occurs rather sharply, then it is reflected in the counting rate of the noble radioactive gas decays. As is known, the ITER plasma temperature will be in the energy equivalent of 10–15 keV, which, according to Eq. (2), will create a Doppler broadening of the neutron pulse in the range 563–790 keV, which

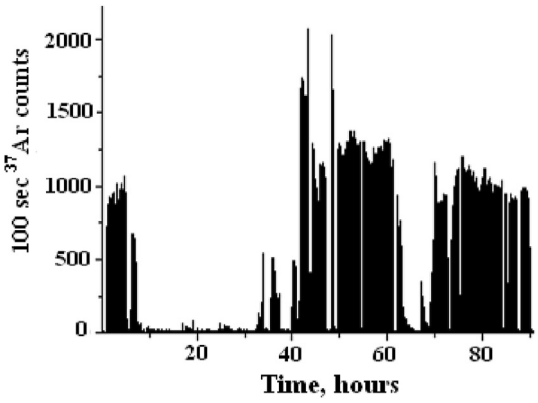

Fig. 4. Illustration of neutron flux monitoring using a radiochemical detector at the installation RADEX.

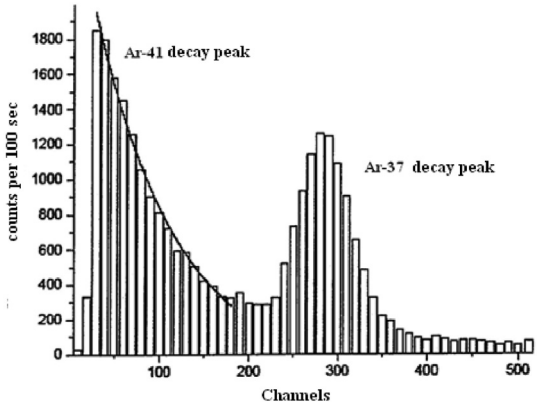

Fig. 5. The decay spectrum on oxalate calcium of natural mixture in a proportional counter.

will significantly change the cross section of the nuclear reaction. As an example, consider the nuclear reaction 11 namely 44Ca (n, α) 41Ar from Table 1. The energy dependence of the cross section of the indicated nuclear reaction is shown in Fig. 6. As can be seen, the energy range of interest 13–15 MeV corresponds to the ascending branch of the cross section dependence, and change in energy in the range $\Delta En = 653–790$ keV corresponds to a change in counting rate of 10–12%. Such a change in counting rate is easily noticeable. Thus, the counter can be calibrated by the counting rate corresponding to the temperature of the thermonuclear plasma. Instead of the indicated reaction 11 from Table 1 it is possible to use the other reactions indicated there, with the exception of reactions

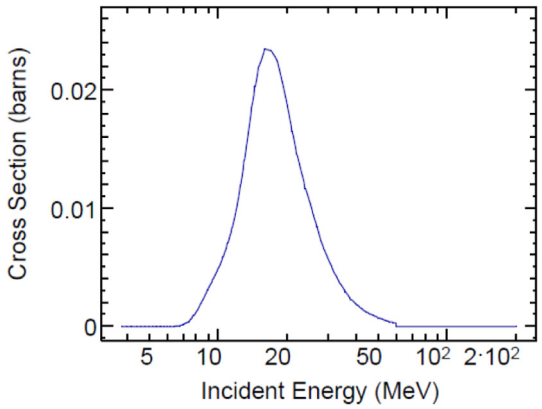

Fig. 6. Cross section for the 44Ca(n,α)41Ar reaction.

with a too short half-life of the radioactive noble gas isotope, which does not allow measurements to be made due to short decay of the corresponding isotope. These include reactions 1–4 and, and to some extent, 5–6 from Table 1. It is interesting to use for thermonuclear plasma thermometry the reaction 9 namely 40Ca (n,α) 37Ar from Table 1. Here, in the energy range 13–15 MeV, the cross section decreases, but in the energy range ΔEn, the change in the cross section is more significant compared to reaction 11 and amounts of 15%. Thus, using reaction 9, it is possible to perform thermometry on the falling branch of the cross section. Moreover in the case of reaction 18 namely 138Ba (n,α)135Xe, the change in the cross section in the interval ΔEn will amount to 25% at all (see Fig. 7). However, it is not clear yet whether the corresponding active substance, barium oxalate, exists and what its properties are. Both reactions 9 and 11, which are favorable for thermometry, were previously used to monitor the neutron flux of the RADEX facility on a natural mixture of calcium isotopes. The corresponding decay spectrum is shown in Fig. 5. As can be seen, the decay count of reaction 11 is one and a half times higher than that of reaction 9, which agrees well with the estimate according to Eq. (1).

3. Multichannel monitor of the neutron flux distribution inside the active zone of a neutron target

The high sensitivity of the GRCM for detecting ionizing radiation, the relatively high thermal stability and low density of the used active substances - oxalates, as well as their insensitivity to radiation damage make it possible to create compact “point-like” detectors for measuring of neutron fluxes inside the active zone of a neutron target. The detector head represents small steel cylinders with active substance. The mass of active substance is selected

Table 2
Characteristics of nuclear reactions involving protons producing noble radioactive gases.

№	NUCLEAR REACTION	ISOTOPE CONTENT IN NATURAL MIXTURE	HALF LIFE OF GAS PRODUCT	REACTION THRESHOLD, MeV	CROSS SECTION AT 14.5 MEV
1	19F(p,n)19Ne	100%	17.2 s.	4.0	70 mB
2	37Cl(p,n)37Ar	24.23%	35 days	1.5	300 mB
3	79Br(p,n)79Kr	50.7%	35 h	4.0	900 mB
4	79Br(p,3n)77Kr	50.7%	74.7 min.	15	250 mB
5	79Br(p,4n)76Kr	50.7%	14.8 h	35	45 mB
6	127I(p,n)127Xe	100%	36.4 days	3.0	500 mB
7	127I(p,3n)125Xe	100%	17.0 h	20	850 mB
8	127I(p,5n)123Xe	100%	2.08 h	40	500 mB
9	127I(p,6n)122Xe	100%	20.1 h	50	125 mB
10	127I(p,7n)121Xe	100%	40.1 min.	55	90 mB

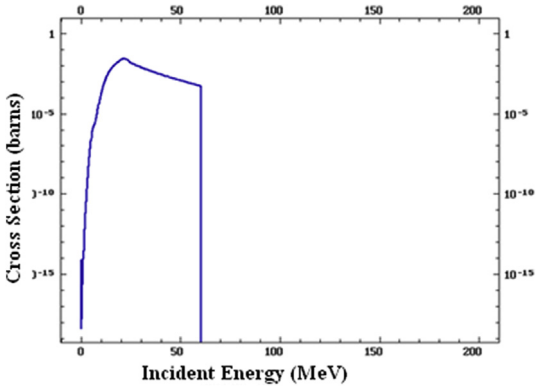

Fig. 7. Cross section of nuclear reaction 138Ba (n, α)135Xe.

based on the magnitude of the neutron flux to ensure an acceptable count rate in the remote counter. By placing several such detectors in different parts of the neutron target, it is possible to obtain the distribution of neutron fluxes over the active zone of the target in real time. Another possible application of multichannel GRCM will be in the neutron target with ion beam driving. In the linear proton accelerator of Moscow Meson Facility [17] of INR RAS the multichannel GRCM can be used for controlling the guidance accuracy of the initiating proton beam to the center of the neutron target. Such a neutron target is the RADEX installation of the INR RAS. Fig. 8 shows a scheme of the gas pipeline of a proposed 5-channel GRCM hodoscope for monitoring of proton beam position on the surface of the neutron target. This detector is supposed to be used as an internal diagnostic tool, as well as for monitoring the distribution of neutron fluxes in the middle section of the neutron core. All five detectors are supposed to be placed crosswise in one plane, as shown in Fig. 8. The proton beam guidance is controlled by comparing the decay counts at the central and peripheral detectors. The technical implementation of such a scheme does not pose a significant difficulty, because the active zone of RADEX facility is a set of tungsten plates, the planes of which are perpendicular to the axis of the initiating proton beam. Hodoscope detector heads and capillary tubes of the gaseous pipeline are supposed to be placed inside a steel dissected plate, the dimensions of which coincide with those of tungsten plates. Thus, a steel plate with hodoscope sensors replaces one of the tungsten plates inside the neutron target core and can be considered as a moveable device, the position of which can be if necessary, changed to study the distribution of neutron fluxes and the shape of the neutron source inside the neutron target core.

The gaseous pipeline of the hodoscope is supposed to be equipped with thin stainless capillary tubes with a bore of about ~1 mm to reduce the delay time between nuclear reaction inside the target core and registration in the remote counter. The tubes are assumed to be brought out through the upper flange of the neutron target casing.

4. GRCM implementation for study of astrophysical objects

An interesting opportunity for the implementation of a GRCM is provided by spectral analysis of luminous bodies. This approach may be useful [3] in the study of astronomical objects.

- to obtain the information about the structure of the object and the location of its parts.
- to obtain the information about the physical processes occurring in them
- to determine the movement of objects and their parts

It is known that information about the elemental composition of stars is contained in their emission spectra. Using spectral analysis, measurements were made of the elemental composition of the Sun and numerous stars, most of which are millions and billions of light years distant from Earth. Moreover, the intensity of the lines of specific elements corresponds to their concentrations. Despite the fact that, according to theory, thermonuclear burning ends with the synthesis of nickel-56 nuclei, the spectra of some stars revealed heavy elements and even radioactive promethium with a half-life of only 17.7 years [18]. In order to detect such a short-lived isotope, one had to carry on longest continuous observations and use powerful computers. And that is important. Indeed, the higher the sensitivity of the detectors, the weaker the spectral lines they distinguish. It turns out a palisade of lines separated by a few angstroms. The fundamental possibility of unstable elements detecting in the spectra of astrophysical objects allows us hopping

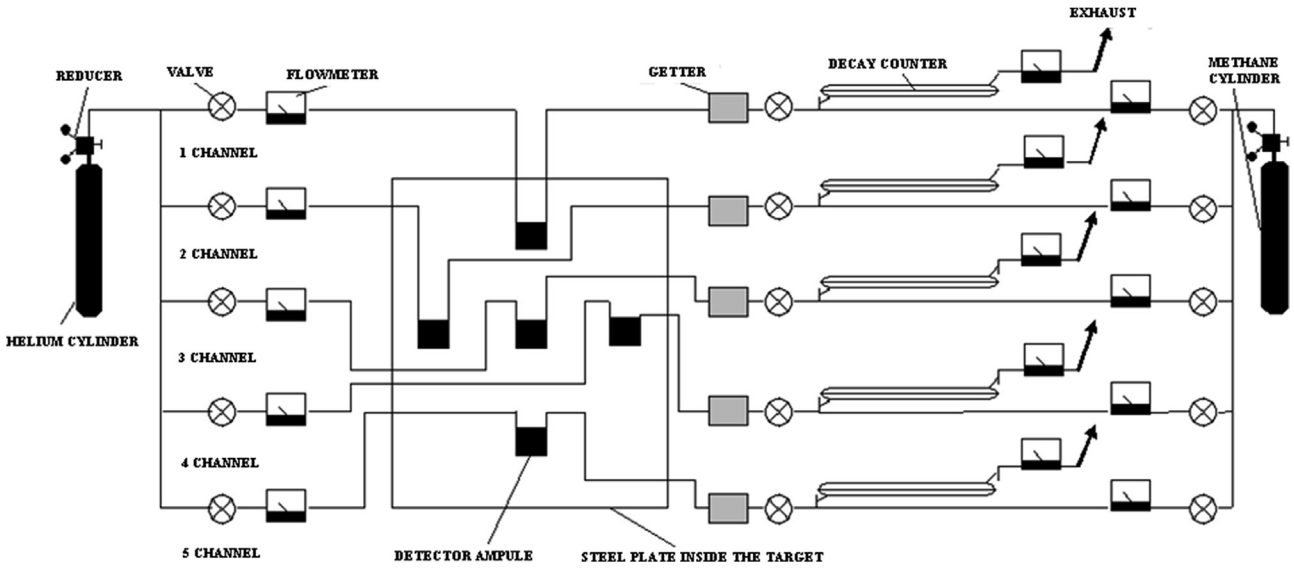

Fig. 8. Scheme of the gaseous pipeline of a 5-channel GRCM hodoscope to control the position of the proton beam on the surface of the RADEX neutron target.

to use GRCM for studying their structure and dynamic changes.

It is known that in the structure of the Sun there are several regions of different densities (see Fig. 9). A core with a density of about 150 g cm-3, where nuclear fusion reactions take place, a radiative transfer zone with a maximum density of 20 g cm-3, and about 30% of the Sun radius is occupied by a convective transfer zone. Here it is believed the nuclear reactions of production of noble radioactive gases and their transfer may occur. The presence of noble radioactive gases can be identified by spectral analysis methods pointing above. The presence of stable isotopes of inert gases such as neon and argon is recognized in the spectrum of the Sun [19]. It is believed that such relatively heavy elements are produced by threshold nuclear reaction in convection zone. The advantage of unstable isotopes of noble gases lies in the possibility to determine the places of their production and travel path inside the Sun and other astrophysical objects, which allows studying the structure and dynamics of the corresponding astrophysical objects. Among the most interesting radioactive isotopes of noble gases such as 39Ar with a half-life t1/2 = 269 years, 42Ar (t1/2 = 33 years), 35Ar (t1/2 = 35 days), 81Kr (t1/2 = 2 × 105 years), 85Kr (t1/2 = 11 years), 79Kr (t1/2 = 35 h), 127Xe (t1/2 = 36 days), 122Xe (t1/2 = 20 h), 125Xe (t1/2 = 17 h), 222Rn (t1/2 = 4 days), 211Rn (t1/2 = 15 h) can be noted. The above list presents isotopes, observations of which allow to study the fluxes of initiating particles, types of nuclear reactions and their places of origin in the atmospheres of astrophysical objects.

The use of the GRCM technique for studying processes in the sun can be illustrated by the following example. Sun flare neutrons are generated at considerable depths in the solar photosphere, where the density of matter exceeds the value of 1014 cm-3 [20–22]. The exact place of neutron generation in the sun's atmosphere has not yet been established. It is possible to clarify the location of neutron sources using radioactive noble gases. There is the reason to believe that noble radioactive gases are generated in the atmosphere of the sun [19] during the interaction of flare neutrons with the corresponding elements [23,24]. Since the spectral lines of noble radioactive gases are observed only during their lifetime, it is necessary to continuously monitor the solar spectrum in order to detect such "flashing" lines. According to the available data on solar flares [25], directly related to the generation of neutrons, the most probable region of their production is within the convective zone. In the convection area with a total height of about 200,000 km the speed of movement of solar matter is 1–2 km/s, while in the chromosphere and photosphere it is much higher and amounts to hundreds of thousands of km/s. Therefore, the localization of neutron sources can be determined using fairly short-lived isotopes

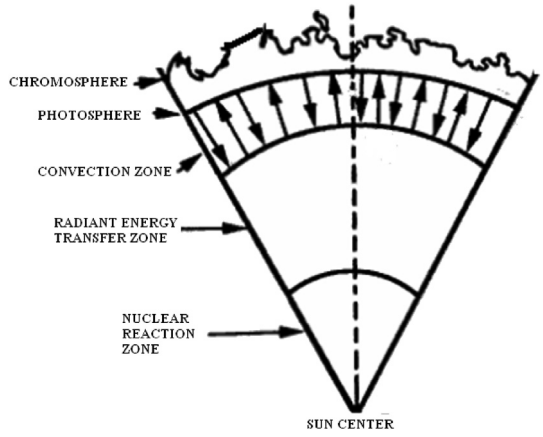

Fig. 9. The structure of the sun.

of radioactive noble gases with a half-life of less than 20 days. However, there is the possibility of testing deeper regions of the sun's atmosphere using long-lived isotopes of noble radioactive gases such as for example 85Kr or even 39Ar. As shown in Ref. [26], the time profile of flare neutron generation turns out to be identical to the profile of gamma radiation in the energy range 4–7 MeV. This fact is favorable for the threshold nuclear reactions of Table 1. A similar conclusion can be made regarding the threshold nuclear reactions initiated by protons, presented in Table 2. With the help of these reactions, by analogy with neutrons, it is possible to determine the localization of the production centers of flare protons. For short-lived radioactive isotopes of noble gases, the location of the fast neutrons and protons sources can be determined using the decay time of the spectral line initial intensity of the selected isotope. As is known, the time dependence of the isotope intensity has the form

$$I(t) \cdot = \cdot I0_{exp}\ (-t \cdot / \cdot t1/2), \tag{3}$$

where I0 is the initial intensity of the isotope line at the formation in a source of fast neutrons or protons, t is the decay time of a radioactive isotope as it diffuses into the photosphere. Let us denote the time of appearance of the isotope spectral line t1. After a time t2 equal to several half-lives, the isotope line in the sun's spectrum will disappear. Let us compose the ratio of line intensities at times t1 and t2

$$K = \exp\{(t2 - t1)/t1/2\}, \tag{4}$$

Then, for the thickness of the atmospheric layer separating the isotope birthplace and the spectral line emission site in the photosphere, we obtain the expression

$$h = vt1 = v\ (t2 - t1/\ 2\ln K), \tag{5}$$

where v is the movement rate of the atoms of the radioactive isotope in the atmosphere of the sun. Of course, the indicated Eqs. (3)–(5) can be used only for the estimated determination of the location of neutron and proton sources; however, on the basis of the proposed simple approach a more accurate calculation can be developed.

6. Radiochemical detector of the spatial distribution of the fast neutron flux density in a nuclear reactor

Measurement of the distribution of the fast neutron flux density (DFND) along the height of the fuel element operating in full power reactor mode allows estimating the corresponding distribution of the degree of fuel burnup. This kind of information makes it possible to use nuclear fuel more efficiently. However, on-line measurement of DFND in power reactors when operating at full power is extremely difficult, primarily due to the lack of adequate means of measuring of DFND in the fast scan mode along the height of the fuel elements in the condition of high neutron flux value of Fn ~1013 cm-2sec-1 and higher. However the DFND may be possible thanks to GRCM. As the active substance for such detector anhydrous calcium oxalate powder CaC2O4 as in early cases [1–4] was proposed [27].

Upon transition to high neutron fluxes Fn ≥ 1011 cm-2sec-1, it becomes possible to use an extended target in the form of a chromatographic column placed, for example, in the technological channel of a nuclear reactor fuel assembly. Such a column should be filled with helium [4,27]. The scheme of the proposed device is presented in Fig. 10. The ampoule 1, made in the form of a long column and placed in a neutron flux, contains a powdered active substance, which is the microcrystalline powder of anhydrous

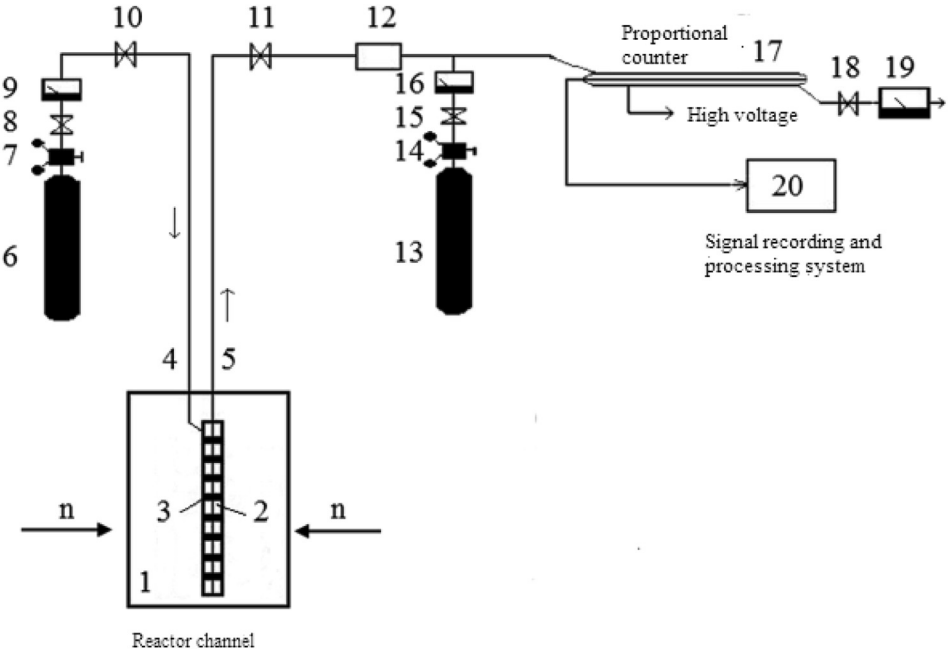

Fig. 10. Scheme of a radiochemical detector of spatial distribution of the fast neutron flux in a nuclear reactor.

calcium oxalate CaC2O4. The active substance is divided into domains 2, separated by equal intervals, in which extended porous inserts 3 of inert material are placed. The input of the ampoule 4 is connected to a reservoir 6 with a gas - carrier, and the output 5 is connected to the input of a flow counter 17 and a signal recording and processing system 20. The gas - carrier from the reservoir (cylinder) 6 through the reducer 7, the flow regulator 8, the flow meter 9 and the valve 10 passes through the ampoule 1, entraining the noble radioactive gas 37Ar formed in the reaction 40Ca(n, α) 37Ar, and then through the valve 11 and getter 12 passes into the flow counter 17. After counter 17, gas passes through the valve 18 and flow meter 19 and exits the registration system. DFND allows realizing the transformation of longitudinal coordinate distribution of the argon isotopes activity along the chromatographic column into a temporal distribution of the decay rate of the counter. Such counting rate temporal distribution can be considered as peculiar "barcode" allowing seeing an instant picture of the neutron flux distribution along the height of the active zone. In more detail DFND and the principle of its work is described in Ref. [4].

5. Discussion

GRCM is characterized by the presence of an intermediate agent, a gas-carrier, transporting radioactive gas product of a nuclear reaction from the production zone to the decay counter. Such functional dividing can significantly reduce the background load in the counter due to the uniqueness of the nuclear reaction, the product of which is a well-defined radioactive noble gas. In addition, the removal of the decay counter from zone of nuclear reactions and its placement inside the biological shielding allows minimizing the effect of background events on the measurement results. The use of noble gas as a carrier and radioactive product of nuclear reactions avoids the interaction of these gases with the structural materials of the gas pipeline. The disadvantage of a flow-through gas method for detecting of threshold nuclear reactions is the time delay of the signal in the counter relative to the moments of nuclear reactions in the core. This problem does not allow the use of this method in situations where a quick control effect on processes in the core is necessary. In addition to neutrons, the GRCM makes it possible to detect protons, deuterons, tritons, and possibly heavier nuclei if their energies are high enough to initiate the nuclear reactions with production of noble radioactive gases.

The thermonuclear plasma thermometry method proposed in the article is based on the threshold behavior of (n, α), (n, p) and similar nuclear reactions just in the desired energy range of about 14 MeV. Due to the existing uncertainties in the cross sections values of these nuclear reactions, similar method can be considered as evaluative and can be used as additional independent information about the plasma temperature.

The radiochemical detector of spatial distribution of the fast neutron flux density in a nuclear reactor works on the same principle as its prototype - GRCM neutron detector, therefore its operability is beyond the doubt. The novelty of the proposal is due to an important new area of application - the measurement of the distribution of neutron flux density over the height of the active zone of a nuclear reactor. DFND allows realizing the transformation of longitudinal coordinate distribution of the argon isotopes activity along the chromatographic column into a temporal distribution of the counting rate of the detector. Such counting rate temporal distribution can be considered as peculiar "barcode". Thus, instead of measuring the neutron flux at one point of the prototype, a simultaneous measurement of the neutron flux at several points along the vertical axis of the active zone of a nuclear reactor can be performed. In this case, all the conditions necessary for the reliable operation of the device are met: the neutron flux density is much higher than the value at which the prototype worked confidently, which ensures reliable registration. Cooling of the device is ensured by the flow of gas - carrier, as well as by the staff cooling system of the fuel assembly, which guarantees the operability of the device in the working temperature range $T < 380\ ^{\circ}C$.

The fundamental possibility of unstable elements detecting in the spectra of astrophysical objects allows hopping to use the GRCM to study their structure and dynamic changes. The advantage of unstable isotopes of noble gases lies in the possibility of determining the places of their production and distances of movement inside the stars and other astrophysical objects.

A multichannel monitor of the neutron fluxes distribution inside the neutron target core allows precise guidance of the particle beam on the neutron target, which is relevant for neutron targets with an external driver.

6. Conclusion

Despite of its insufficient fame, the gaseous radiochemical method for radiation detection has broad prospects for application in such areas as neutrino detection, monitoring of neutron fluxes in nuclear facilities, diagnostics of charged particles, thermometry of thermonuclear plasma, study of the structure and dynamics of astronomical objects, position-sensitive dosimetry, and measurement of spectra of bombarding particles. The advantages of GRCM are insensitivity to gamma quanta due to the possibility of spatial separation of the detector and decay counter, the simplicity of the detection ampoule and the absence of moving parts. The use of inert gases as a carrier and a decaying agent allow avoiding any interaction of the gas flow with structural elements in the gas pipeline and the decay counter. The use of GRCM for monitoring neutrons in high-flux nuclear and thermonuclear installations, where the measurement limit of the neutron flux density is determined only by the count rate of the information collection and processing system, seems especially promising. A fresh proposal is the possibility of GRCM using in the study of the structure and dynamics of astrophysical objects. Success in this direction may be the result of the development of spectral analysis with the aim of detecting of "flashing" lines of radioactive isotopes of noble gases in the spectra of astrophysical objects.

Declaration of competing interest

We wish to that there are no conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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