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Monitoring of Neutrons Field of the WWR-SM Reactor at Reception of Phosphorus-33

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Abstract In the paper results of monitoring of thermal neutrons field in vertical irradiation channels of the WWR-SM nuclear research reactor of INP AS, Tashkent, Uzbekistan has been studied with purpose of determination of mode of irradiation target sulfur-33 (S-33) and increase specific activity of phosphorus-33 (P-33). It is shown that reception P-33 with specific activity (120-140 mCi/g) is necessary to spend irradiation targets of S-33 elemental enrichment in vertical channels of the WWR-SM reactor in the following mode: irradiation place is distance of 35-50 cm below from top point of the vertical channel, nominal power of reactor=10MW, reactor thermal neutrons stream density $\geq 0.7 \cdot 10^{14}$ neutrons/cm^2·sec and irradiation time=320÷620 hours. Opportunity of application of S-33 as the monitor for reception of P-33 without the carrier was established. In fist time for express definition of specific activity of radionuclide P-33 in irradiation target S-33 the monitor S-33 was used.

Keywords Nuclear Reactor, Thermal Neutrons, Thermo-neutron Gauge, Irradiation Target, Monitoring of Neutrons Field, Output of Phosphorus-33

1. Introduction

The radionuclide P-33 having half-life period ($T_{1/2}$= 25.4 days), average energy beta-radiation E_β=77 keV and maximal energy beta-radiation 243 keV is widely used in nuclear medicine and genetic engineering. For carrying out biological and medical scientific researches, production of radionuclide P-33 with high specific activity is important [1-3].

The radionuclide P-33 is received at irradiation at an irradiation neutrons of a reactor of a target of S-33 on exoenergy nuclear reaction ^{33}S (n, p) ^{33}P, therefore at an irradiation of S-33 the share radionuclide P-33 increases with reduction of energy of neutrons [4].

For reception radionuclide P-33 without carrier use radiochemical processing of the irradiate target of sulfur, enriched elemental ^{33}S isotope, which has high cost (~$ 200 per milligram for 90% enrichment) [5].

In nuclear reactor for reception radionuclide P-33 high specific activity it is necessary to increase thermal neutrons stream density. In WWR-SM reactor this problem can be solved at use fuel assembly with High-Enrichment Uranium (HEU) IRT-3M fuel with 90% U-235 enrichment or IRT-3M fuel with 36% U-235 enrichment at nominal power of reactor 10 MW and high streams of thermal neutrons. However, the WWR-SM reactor on Low-Enrichment Uranium (LEU) fuel of type IRT-4M with 19.7% U-235 enrichment was conversed, as according to the Reduced Enrichment Research and Test Reactor (RERTR) Program established in 1978 at the Argonne National Laboratory USA by the Department of Energy, USA [6-8]. In these conditions at use LEU fuel in vertical irradiation channels of WWR-SM reactor to reach high streams of thermal neutrons is practically impossible, hence for increase in specific activity P-33 optimization of mode of irradiation of target S-33 is required.

The purpose research is monitoring thermal neutrons field of WWR-SM reactor, optimization of mode of neutron irradiation of target S-33 in vertical channels of WWR-SM reactor for increase specific activity of radionuclide P-33.

For decision of this problem researches on monitoring thermal neutrons field of the WWR-SM reactor, distribution of thermal neutrons stream density lengthways vertical channel and opportunity to use as the monitor of S-33 for express definition of specific activity of radionuclide P-33 in irradiated target S-33 have been studied.

2. Materials and Methods

Usually in low power reactzors (including WWR-SM reactor) for measurement of thermal neutrons densities monitors Co-59, Na-24, Mo-98, Dy-164, In-113, In-115, Mn-55, Cu-63, Cu-65, Au-197 [9] or monocrystalline silicon [10] can be used.

Monitors (detectors) disks (foil) alloyed cobalt with aluminum with the content of Co-59 (0.1 %) in diameter of 3.0 mm and thickness of $4.4\cdot10^{16}$ kernels/cm^2 with cadmium screens of two types CE-1 (thickness 0.5 mm) and CE-2 (thickness 1.0 mm) was used. Detectors of Co-59 has the following nuclear-physical characteristics: isotopic concentration=100%, type of nuclear reaction ^{59}Co (n ,ϒ) ^{60}Co, cross section for thermal neutrons σ=37.35 barn, decay constant=4.16910^{-9}sec^{-1}, half-life of Co-60=46182.43 hour, output of gamma-quantum $R_ϒ$=0.998, energy of gamma-quantum $E_ϒ$=1332 keV, thermal neutrons energy $E_n \leq 0.025$ eV, time of endurance of irradiated target=48÷72 hours.

Monitors of S-33 (weight=1.0 mg) have been placed in quartz ampoule (diameter=2.0 mm and length=10÷15 mm). Then monitors of Co-59 (cobalt alloy with Co-59 0.1%) in form of metal disks (∅=3.0 mm, h=0.2 mm, m=3.0 mg) together with monitors of S-33 in quartz ampoule have been placed in the aluminum block container and in vertical channel of WWR-SM reactor in current estimated time have been irradiated. After, irradiated monitor of S-33 in small volume (2-3 ml) of organic solvent n-hexane is dissolved and value activity of formed radionuclide P-33 specific activity is defined by measuring of aliquot part radioactive sulfur solution. Measurement of the induced activity of radionuclide Co-60 and aliquot part sulfur solution with radionuclide P-33 in the beta-gamma-spectrometer «Progress BG (II)» BDEB3-2U with the software “Progress5” have been spent. Also measurement of induced activity of radionuclide Co-60 in cobalt monitor (after irradiation monitors held out during 3-4 days) on multichannel gamma-spectrometer SU-01P with Ge-Li detector DGDK-100 with the software «Aspekt, Angamma» has been spent and activity of monitor Co-60 on gamma-spectrum Co-60 with gamma-energy $E_ϒ$=1332.5 keV has been defined.

In the beginning defined thermal neutrons stream density by using method of neutron activation of Co-59 metal disks (foil) has been spent, then activity radionuclide P-33 have been executed.

Activity of radioniclide Co-60 in the monitor Co-59 was defined at use the formula [11]:

$$A=\frac{0,6\cdot\theta\cdot\Phi\cdot m\cdot\delta\cdot(1-e^{-\frac{0,693\cdot t_0}{T_{1/2}}})\cdot(1-e^{-\frac{0,693\cdot t_1}{T_{1/2}}})}{M\cdot 3,7\cdot10^7} \quad (1)$$

where: A – activity of Co-60, mCi; Φ - thermal neutrons stream density, neutrons/cm^{2}sec; t_0 - irradiation time of sample, sec; t_1 - held out time of irradiated sample, sec; $T_{1/2}$ - half-life of Co-60, sec; θ - enrichment of Co-59, %; m - weight of Co-60 target, g; δ - activation cross section of nuclear reaction ^{59}Co(n,ϒ)^{60}Co, barn.

Thermal neutrons stream density was defined at use the formula [11]:

$$\Phi = \frac{A\cdot e^{\lambda t}}{N\cdot\sigma\cdot(1-e^{-\lambda t_0})} \quad (2)$$

Φ – thermal neutrons stream density, neutrons/cm^{2}sec; A — activity of radionuclide Co-60 in monitor Co-59, imp/sec (photopic area); N – number of kernels Co-59; t - time of endurance detector, sec; σ - cross section of Co-60, barn; t_0 – irradiation time of monitor, sec; λ - decay constant of Co-60; $T_{½}$ - half-life of Co-60, sec.

Shown under the formula 2N – number of kernels of Co-59 is defined under the formula [11]:

$$N = (m\cdot N_A\cdot k\cdot\eta)/A \quad (3)$$

where: m - weight of the irradiated detector, g; N_A - Avogadro's constant ($N_A=6,02\cdot10^{23}$); k - prevalence of isotope; η-share of atoms of the detector in an irradiated target; A - nuclear weight of isotope Co-59, g.

Activity of radionuclide P-33 in the target S-33 has been certain under the formula [11]:

$$A=\frac{0,6\cdot\theta\cdot\Phi\cdot m\cdot\delta\cdot(1-e^{-\frac{0,693\cdot t_0}{T_{1/2}}})\cdot(1-e^{-\frac{0,693\cdot t_1}{T_{1/2}}})}{M\cdot 3,7\cdot10^7} \quad (4)$$

where: A – activity of radionuclide P-33, mCi; Φ – thermal neutrons stream density, neutrons/cm^{2}sec; t_0 – irradiation time of sample, sec; t_1 – held out time of irradiated sample, sec; $T_{1/2}$ - half-life of P-33, sec; θ - enrichment of S- 33,%; m – weight of target S-33, g; δ - activation cross section of nuclear reaction ^{33}S (n, p)^{33}P, barn.

For study in distribution of thermal neutrons stream density in length ways of vertical channel of WWR-SM reactor thermo-neutron gauge TND-2.0 was used. TND-2.0 operates in the range from $5\cdot10^{12}$ neutrons/cm$^2\cdot$sec to $5\cdot10^{14}$ neutrons/cm$^2\cdot$sec in gas environments and in water. The principle work of thermo-neutron gauge TND-2,0 [12] consists on influence of neutrons on sensitive element of the neutron detector which structure includes sharing under action of neutrons material uranium-nickel an alloy and differential thermo couples alloy (chromel and alumel). At action of pulse neutron irradiation on sensitive element TND-2.0 there is transformation of nuclear reactions energy of uranium division to thermal energy which part will be transformed to electric energy by means of the differential thermo couples which are being thermal contact to sensitive element.

The following measurement technique of potential difference in vertical channels of reactor has been used: aluminum bar (length=6.0m) with attached on the end thermo-neutron gauge TND-2.0 was lowered inside of the vertical channel, fixed on distance 5.0 cm in direction from top to the bottom of the vertical channel, at reactor low power (300 kW) and short-term irradiation time (15-30 minutes) in this point the potential difference has been measured with use secondary device potentiometer P-4833 (connected through electric wires with TND-2.0) which is located on protective cover of the rector. Then the thermo-neutron gauge TND-2.0 has been lowered downwards on 5.0 cm, it is anew fixed and was measured potential difference and so on. Further on the point of measurement where was determined the highest potential difference neutron activation monitor of Co-59 is attached on the aluminum bar end and it`s irradiation was spent in same regime. For determination of activity of radionuclide Co-60 the multi-channel gamma-spectrometer SU-01P with Ge-Li detector DGDK-100 with the software «Aspekt, Angamma» have been used.

In the vertical channel of reactor monitors Co-60 and S-33 together with target sulfur have been irradiated by neutrons in the block container EK-10 (length is 340 mm and diameter is 25 mm) [13], which is installed in the central hollow part of 6-trumpet fuel assemble type of IRT-4M with low enrichment fuel (19.7% on U-235).

3. Results and Discussions

In the Figure 1 constructional scheme of the 6-trumpet fuel assembly type of IRT-4M is shown. Fuel assemble type of IRT-4M has hollow empty part, where target of S-33 together with monitors Co-59 and S-33 in the special block container EK-10 was loaded in active core of WWR-SM reactor.

In Table 1 in vertical channels of reactor WWR-SM values of potential differences measured by thermo-neutron gauge TND-2.0 and potentiometer P-4833 are shown.

The results of thermal neutrons stream density was measured on height of the vertical channel 3-4 at use HEU fuel assembly type of IRT-3M received in 1986 are shown in table 2.

Based on table 1 dependences of the potential differences on points of measurements along vertical channels has been made. Curves dependences of the potential differences on distances from top to bottom of vertical channels of WWR-SM reactor are shown in Figure 2.

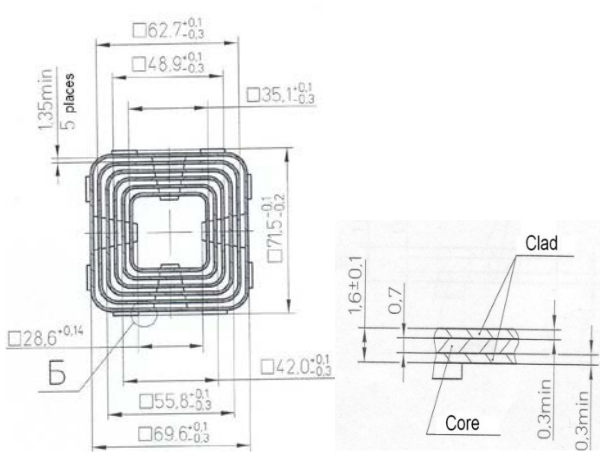

Figure 1. Drawing of 6-trumpet fuel assembly tape of IRT-4M with central hollow part (top view). Б – core shell with UO^2- Al.

Table 1. Potential difference in regular vertical channels of the reactor WWR-SM (reactor power=10MW)

Distance below of the top point of the vertical channel, cm	Number of the vertical channel					
	5-7	4-4	4-2	3-4	3-7	2
	Potential difference, mV					
10.0	8.0	11.1	10.3	10.,0	10.0	6.0
15.0	10.2	11.1	14.2	11.1	12.6	8.0
20.0	13.7	14.1	18.,5	13.9	16.,4	10.4
25.0	16.2	17.1	22.0	16.7	19.,8	12.1
30.0	18.4	19.8	25.,3	19.,2	22.1	13.5
35.0	20.0	21.3	27.,0	20.5	23.8	14.1
40.0	21.1	22.0	28.,6	21.2	25.,0	14.1
45.0	20.0	22.0	28.0	21.0	24.4	14.1
50.0	18.6	21,0	25.8	20.0	23.0	13.0
55.0	16.0	18.8	23.0	19.0	20.0	12,0
60.0	15.5	15.2	22.0	16.4	19.,9	11.6
65.0	13.,2	13.,0	15.4	14.2	16.2	10.2

Table 2. Valuesof thermal neutrons stream density on height of the channel 3-4 of active core

Height, mm	Thermal neutrons stream density, neutrons/cm²sec	Height, mm	Thermal neutrons stream density, neutrons/cm²sec
6900*	$0.16{\cdot}10^{12}$	6300***	$1.91{\cdot}10^{13}$
6850	$0.23{\cdot}10^{12}$	6850	$1.86{\cdot}10^{13}$
6800	$0.48{\cdot}10^{12}$	6800	$1.75{\cdot}10^{13}$
6750	$0.13{\cdot}10^{12}$	6750	$1.6{\cdot}10^{13}$
6700	$2.34{\cdot}10^{12}$	6700	$1.34{\cdot}10^{13}$
6650	$4.58{\cdot}10^{12}$	6650	$1.06{\cdot}10^{13}$
6600	$7.26{\cdot}10^{12}$	6600	$7.48{\cdot}10^{12}$
6550	$1.02{\cdot}10^{13}$	6550	$4,69{\cdot}10^{12}$
6500	$0.14{\cdot}10^{13}$	6500	$2,54{\cdot}10^{12}$
6450	$0.65{\cdot}10^{13}$	6450	$1.35{\cdot}10^{12}$
6400	$0.8{\cdot}10^{13}$	6400	$1.25{\cdot}10^{12}$
6350**	$0.88{\cdot}10^{13}$		

*bottom of irradiated vertical channel. **physical center of reactor core. ***geometrical center of reactor core.

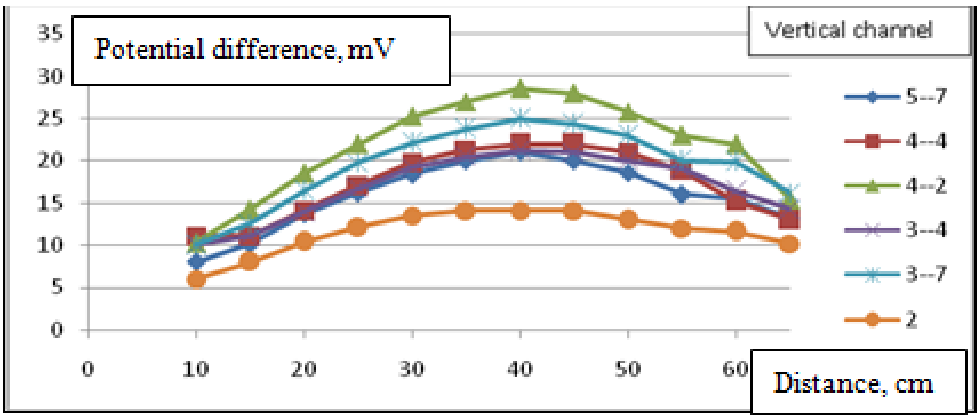

Figure 2. Curves dependences of potentials differences on distances from top to bottom of vertical irradiation channels

Thus, in irradiation vertical channel of WWR-SM reactor values the potentials differences and, correspondingly, the thermal neutrons stream density has high values at distance of 35-50 cm from the top point of the vertical channel, therefore irradiation of target S-33 was provided on this interval distance of the vertical channel. The results shown in table 1 will well be agreed with the results received in 1986, when in active core of WWR-SM reactor HEU fuel assembly type of IRT-3M with 36% U-235 enrichment was used (Table2).

Influence of value of thermal neutrons stream density on output specific activity of radionuclide P-33 has been studied. With this purpose in current 120 hours irradiation of target S-33 in the peripheral vertical channel 5-1 has been spent, where the thermal neutrons stream density twice is less in comparison with thermal neutrons stream density in vertical channels located in the center of active core. After processing irradiation of the raw material S-33 the output of P-33 was made practically 100% from the calculated value of the induced activity of P-33. At reactor power 5.0 MW the thermal neutrons stream density in the central channels corresponded $1.0{\cdot}10^{14}$ neutrons/cm$^2{\cdot}$sec and output of the induced activity of radionuclide P-33 corresponded to value 500 mCi/g $_{(S\text{-}33)}$ at irradiation of target S-33 during 1000 hours, and 320 mCi/g $_{(S\text{-}33)}$ at irradiation of target during 500 hours. Hence, for reception of radionuclide P-33 without damage it is possible to spend irradiation of target S-33 at the lowered power of WWR-SM reactor up to 5.0 MW.

Monitoring of the thermal neutrons stream density and definition of output value of induced activity of radionuclide P-33 received at irradiation S-33 in vertical channels of WWR-SM reactor at use fuel assembly of type IRT-3M with 90% enrichment on U-235 executed in 1994 is shown in table 3.

Activity of radionuclide P-33 calculated and practical output of induced activity of radionuclide P-33 during estimated irradiation time are shown in table 4.

Monitoring results of neutrons field and definition of value output of induced activity of radionuclide P-33 at irradiation of target S-33 in the vertical channels of reactor at use fuel assembly type of IRT-4M with 19.7% enrichment on U-235 carried out in 2017 is shown in table 5.

Table 3. Value of thermal neutrons stream density in the vertical channels

Weight of monitor of Co-59, mg	Irradiation time, hour	Number of the vertical channel	Thermal neutrons stream density average, n/cm^2sec
1.7	48	2-4	$1.3 \cdot 10^{14}$
3.1	50	5-5	$6.9 \cdot 10^{13}$
2.1	50	2-5	$1.3 \cdot 10^{14}$

Table 4. Results of monitoring neutrons fields and definition value of output of the induced activity of radionuclide P-33

#	Weight of target S-33, g	Irradiation time of target, hour	Number of vertical channel	Thermal neutrons stream density, $n/cm^2 \cdot s$	Calculated activity of P-33, mCi/g	Practical activity of P-33, mCi/g
1.	6,0	1139	Central 1	$0.31 \cdot 10^{14}$	657	410
2.	6,0	1129.8	Central 2	$0.43 \cdot 10^{14}$	736	450
3.	6,0	634.0	Central 3	$0.36 \cdot 10^{14}$	516	430
4.	6,0	1590.3	Central 4	$0.33 \cdot 10^{14}$	760	460

Table 5. Results of monitoring neutrons fields and definition value of output of induced activity of radionuclide P-33.

#	Weight of target S-33, g	Irradiation time, hour	Number of vertical channel	Thermal neutrons stream density, $n/cm^2 \cdot sec$	Calculated activity of P-33, mCi/g	Practical activity of P-33, mCi/g
1.	4.0	1391	3-2	$0.47 \cdot 10^{14}$	229	214
2.	4.4	1391	2-6	$0.45 \cdot 10^{14}$	220	210
3.	5.0	623	4-2	$0.7 \cdot 10^{14}$	129	121
4.	5.0	321	3-3	$0.8 \cdot 10^{14}$	152	140

From the table 5 it is visible, that values of specific activity of radionuclide P-33 executed at calculation, and practical outputs of radionuclide P-33 will well be coordinated among themselves and in most cases, the practical output of radionuclide P-33 corresponds 50-70% from the calculated value. Practical output of radionuclide P-33 depends on irradiation time of target S-33 and loading irradiation place of target in reactor vertical channel, so, at long irradiation time (1500÷2000 hours) of target S-33 output of radionuclide P-33 is lower, than output of radionuclide P-33 received at continuous irradiation of target S-33 (~500 hours).

In addition, cadmium attitude of radionuclide P-33 has been studied also. For definition cadmium attitude of radionuclide P-33 was used the formula [14]:

$$R_{Cd} = \frac{A_1}{A_2} - 1 \qquad (5)$$

where: R_{Cd} – cadmium attitude, A_1 – activity of radionuclide P-33 at irradiation of target S-33 without cadmium screen, A_2 – activity of radionuclide P-33 at irradiation target S-33 with cadmium screen.

From formula 3 is follows, that the greater the cadmium ratio, the smaller the fraction of resonant neutrons in the total neutron flux density and vice versa, as target S-33 wrapped up in cadmium screen is badly activated by thermal neutrons as cadmium absorbs practically all thermal neutrons, because cross section cadmium has very high value (~2450 barn).

Cadmium attitude for radionuclide P-33 makes ~7.5÷8.0 and it means, that resonant neutrons ($E_n \leq 3.0$ MeV) and fast neutrons ($E_n \geq 3.0$ MeV) make very small partial contribution to the yield of induced activity of radionuclide P-33, and basically activity of radionuclide P-33 increases with increase in thermal neutrons stream density.

4. Conclusions

For reception high specific activity of radionuclide P-33 (120-140 mCi/g) in active core of WWR-SM reactor, the optimum mode of irradiation of target S-33 has been found that process irradiation of quartz ampoule filled with target of S-33 is necessary provide into internal cavity of fuel assemble type of IRT-4M in special block container EK-10, on distance 35-50 cm from top point of vertical channel, at reactor nominal power=10MW, thermal neutrons stream density≥$0,7 \cdot 10^{14}$ neutrons/$cm^2 \cdot sec$ and irradiation time=320÷620 hours.

In fist time for reception of radionuclide P-33 without carrier from irradiated target S-33 opportunity of application as the monitor S-33 was established. Monitor S-33 has some advantages in front of other monitors. Technique of definition of induced activity of monitor S-33 is very simple also does not demand complex difficult radiochemical operations, i.e. the irradiated monitor S-33 very easily is dissolved in small volume of organic solvent n-hexane and value of specific activity of radionuclide P-33 is defined with measuring of aliquot part of radioactive solution of S-33. On activity of radionuclide P-33 in the monitor S-33 it is possible easily calculate common activity of radionuclide P-33 in irradiated target S-33. In manufacture of radionuclide P-33 without carrier the method of control irradiation target S-33 with use monitor S-33 is simply and economic [15]. Knowing the induced activity of radionuclide P-33 in the monitor S-33, it is possible easily determined how much necessary to take targets of S-33 enriched for it is irradiation in vertical channel and reception of necessary activity of radionuclide P-33.

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