Peertechz

ENGINEERING GROUP

Insights in
Nuclear Energy, Science, and Engineering
OPEN ACCESS JOURNAL

DOI: https://dx.doi.org/10.17352/inese

Research Article

Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds

Ulugbek T Ashrapov*, Shavkat R Malikov and Bakhodir B Mirzaev

Department of Nuclear Energy and Nuclear Technologies, Institute of Nuclear Physics, Academy of Sciences, Tashkent, Republic of Uzbekistan

Received: 09 July, 2025
Accepted: 21 July, 2025
Published: 22 July, 2025

***Corresponding author:** Ulugbek T Ashrapov, Department of Nuclear Energy and Nuclear Technologies, Institute of Nuclear Physics of Academy Sciences, Tashkent, Republic of Uzbekistan,
E-mail: ashrapov@inp.uz

Keywords: Sulfur-32; Carbon tetrachloride; Irradiation; Thermal neutrons; Activity; Yield of sulfur-35

https://www.engineegroup.com

Check for updates

Abstract

The article presents methods for obtaining sulfur-35 radionuclide without a carrier from neutron-irradiated potassium chloride, sodium chloride, magnesium chloride, and carbon tetrachloride.The methods of irradiation of targets from chlorine-containing compounds with thermal neutrons in the vertical channel of the WWR-SM reactor, methods of processing irradiated targets, and extraction of sulfur-35 without a carrier are presented.The highest yield of sulfur-35 activity per 1 g of chlorine-containing compound (3.312 Ci/g) is achieved by irradiating CCl_4 targets under the following conditions: thermal neutron flux density is ≥ $1 \cdot 10^{14}$ n/cm^{2}sec, irradiation time is 2000 hours, nominal reactor power is 10 MW, and irradiation of a quartz ampoule with the target in a vertical reactor channel with mandatory cooling of the target with running first loop water. The isolation of sulfur-35 without a carrier from irradiated carbon tetrachloride was carried out using the water extraction method, which is the simplest and does not require complex radiotechnological operations.

Introduction

Radionuclide ^{35}S is continuously produced in the stratosphere, from where it is transported to the troposphere or lower atmosphere and finally transported by rain to groundwater. Once meteoric water enters the subsurface, its ^{35}S activity decreases with a half-life of 87.4 days, making ^{35}S a suitable time tracer for investigating the age of groundwater less than a year [1]. Radionuclide sulphur-35 is released into the environment by the UK nuclear industry during the normal operation of its gas-cooled reactors. The gas is in the form of $CO^{35}S$, which can be easily absorbed by vegetation [2].

The radionuclide ^{35}S is used in nuclear medicine, industry (production of semiconductors and laser technology), and scientific research in the field of biochemistry and microbiology as a radioactive label for proteins, since sulfur is a component of some amino acids of proteins (cystine, cysteine, methionine).

The radionuclide ^{35}S (along with the radionuclide ^{32}P) was first used as a radioactive label in the Hershey-Chase experiments. It is common knowledge that bacteriophages are viruses, one of the simplest objects of living nature. Hershey and Chase demonstrated experimentally that phages inject their DNA, not protein, into bacterial cells. It was known that proteins contain oxygen, nitrogen, carbon, and sulfur, while nucleic acids contain oxygen, nitrogen, carbon, and phosphorus. Sulfur is present in proteins but is absent from DNA, while phosphorus, on the contrary, is present in DNA but is absent from proteins. The experimenters obtained two types of radioactively labeled bacteriophages: some contained the radionuclide ^{35}S, others – the radionuclide ^{32}P. The scientists then "planted" (introduced) two groups of viruses onto the bacteria - one with labeled DNA and the other with labeled protein - and separated the bacteria from the rest of the material using a centrifuge and measured the activity of the radioactive label. As a result of the experiment, it was discovered that the radionuclide 32P was in the bacteria, while the radionuclide 35S remained in the environment. The results showed that the phage DNA penetrates the bacterium and serves as genetic material for the production of the bacteriophage, while its protein shell

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

remains outside the bacterium, and when reproducing, the phage does not penetrate the bacterium entirely: it injects its contents into the bacterium, the protein shell remains outside the bacterial cell, and then new phages are formed inside the bacterium.Thus, the Hershey-Chase experiment demonstrated that the carrier of genetic information in cells is not proteins, as previously thought, but DNA [3].

Radionuclide ^{35}S can also be used in tRNA research, where it serves as a precursor for the formation of selenium-containing compounds such as 5-methylaminomethyl-2-selenuridine [4].

The radionuclide ^{35}S is obtained in a nuclear reactor by the nuclear reaction $^{35}Cl(n,p)^{35}S$ during irradiation of organic and inorganic chlorine-containing compounds. There is no reliable information in the literature on the activation cross-section and threshold energy of neutrons.The activation cross section of the above nuclear reaction ranges from 0.35 to 0.575 barns [5,6].

Radionuclide ^{35}S emits only beta particles with a maximum energy of E_β = 0.167 MeV and an average energy E_β = 0.049 MeV [7].

In a nuclear reaction, decay of the sulfur atom yields, $^{35}_{17}Cl$, with the release of an electron and an antineutrino:

$$^{35}_{17}S \rightarrow {}^{35}_{17}Cl + e^- + \bar{\nu}_e \quad (1)$$

The primary concern for individuals working with the sulfur-35 isotope is the potential for internal exposure if the individual contaminates bare skin, accidentally ingests the material, or inhales it as a gas or vapor. The critical organ for most ^{35}S-labeled compounds is the entire body. Urine testing is an effective sampling method to determine whether ^{35}S absorption has occurred.

$Na^{23}SO_4$ enriched in sulfur-35 is the only commercially available feedstock enriched in ^{35}S, and $Na^{23}SO_4$ is sold at a radioactivity level of 1 mCi. The cost of radionuclide sulfur-35, activity is 1 mCi (37 MBq), specific activity is 1050 - 1600Ci (38.8 - 59.2 TBq)/mmol of the sodium sulfate in 1 ml of water, is $ 1,354.00 - $ 1,604.49. In this regard, obtaining radionuclide ^{35}S without a carrier with high specific activity and radiochemical purity from unenriched, cheap natural chlorine-containing compounds is currently an urgent task.

In the WWR-SM nuclear reactor, to obtain the ^{35}S radionuclide with high specific activity, it is necessary to increase the thermal neutron flux density using a fuel assembly with highly enriched uranium (IRT-3M with 36% enrichment in U-235). However, in 2018, at the request of the IAEA, based on the Reduced Enrichment Reactor Research and Testing (RERTR) Program [8,9], the WWR-SM reactor to low-enriched fuel(IRT-4M with 19,7% enrichment in U-235) was converted. Under these conditions, when using LEU fuel in the reactor active core to obtain a carrier-free highly active sulfur-35, it is necessary to select an optimal irradiation modeand selection of the required chlorine-containing chemical compound as anirradiationtarget toincrease the induced activity of the ^{35}S radionuclide.

This study aims to evaluate the induced activity of radionuclide sulfur-35 during irradiation of chlorine-35 targets with thermal neutronsand to obtain high specific activity of sulfur-35 radionuclide during irradiation of chlorine-containing chemical compounds byneutrons of the WWR-SM reactor.

Materials and methods

As targets for irradiation by thermal neutrons in nuclear reactors, various organic and inorganic chlorine compounds and potassium chloride (KCl) are most commonly irradiated.

In the experiments, the neutron activation of the following chlorine-containing compounds was investigated:KCl - potassium chloride 99,9% (special purity); NaCl - sodium chloride 99,9% (chemically pure); $MgCl_2 \cdot 6H_2O$ - magnesium chloride 6-hydrate (clean for analysis); $CC1_4$ - carbon tetrachloride (clean for analysis). All chlorine-containing compounds met the requirements of the state standard [10-13].

Weighed (1.0 g each)amounts of potassium chloride(KCl), sodium chloride(NaCl), magnesium chloride($MgCl_2$),and carbon tetrachloride(CCl_4) were placed in clean quartz ampoules with an internal diameter ∅ = 4 mm and length l = 50 mm, sealed at a temperature of 1600 °C - 1700 °C. They were then placed in aluminum block containers (with a lead load) together with Co-60 tracking monitorsand loaded into the vertical channels of the reactor. Irradiation was carried out from 100 to 2000 hours at a nominal reactor power of 10 MW, and irradiationby thermal neutron flux density from $1 \cdot 10^{13}$ n/cm²sec to $1.5 \cdot 10^{13}$ n/cm²·sec was made.

After irradiation, irradiated block containers with targets and tracking monitors in the reactor's «hot chamber» were opened, and both the thermal neutron flux densities and the activities of the irradiated targets were measured.

Results

Methodology for irradiating chlorine-containing targets along with track monitors at the reactor vertical channels

Usually, in low power reactors (including reactor WWR-SM) for measurement of thermal neutron flux densities, monitors Co-59, Na-24, Mo-98, Dy-164, In-113, In-115, Mn-55, Cu-63, Cu-65, Au-197 [14] or monocrystalline silicon [15] can be used.

We used monitors of Co-59 in the form of metal discs, which are the most readily available. For the determination of thermal neutron flux density tracking monitors in form of metal disks (foil) alloyed cobalt with aluminum with the contents of Co-59 (0,1%) in diameter of 3,0 mm and thickness of $4.4 \cdot 10^{16}$ kernels/cm² with cadmium screens of two types (CE-1 with thickness 0.5 mm and CE-2 with thickness 1.0 mm) was used.

Several batches of monitors were manufactured: Co-59 monitors (cobalt alloy with 0,1% of Co-59) in the form of metal

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

disks ∅ = 3 Mm, h = 0,2 mm, m = 3,0 mg) each separatelywere packed in aluminum foils.Then each of the tracking monitors,together with potassium chloride, sodium chloride, magnesium chloride, and carbon tetrachloride (each mass is 1.0 g),was placedin quartz ampoules, which were sealed at a temperature of 1730 °C. Irradiation of a quartz ampoule with a target and a tracking monitor, packed in a special aluminum block container (L = 750 mm and = 20 mm) which has a 3 mm diameter hole in the center of the bottom and a3 mm diameter 2 hole in the lid for cooling by water of the quartz ampoule with the target. A special aluminum block container is placed in the internal cavity of the 6-pipe fuel assembly of the IRT-4M type in the active core of the reactor WWR-SM.

Induced activity of radionuclide ^{60}Co was determined bya multichannel gamma-spectrometer SU-01P with a Ge-Li detector DGDK-100 with the software «Aspekt, Angamma»and activity of monitor ^{60}Co bygamma spectrum with gamma-energy E = 1332.5 keVwasmade. The efficiency of the detector of the gamma-spectrometer is defined by means of standard Co-60 from a complete set of etalon spectrometric gamma sources (OSGI).

Thermal neutron flux density determination in the irradiation channel by the following formula was made:

$$F_{th} = \frac{A \cdot e^{\lambda \cdot t_1}}{N \cdot \sigma \cdot (1 - e^{-\lambda \cdot t_0})} \quad (1)$$

Where:

F_t – thermal neutrons flux density, neutrons/cm²·sec;

A – measured activity of the monitor, imp/sec;

N – number of kernels Co-59 (4.4·10^{16} kernels/cm²);

t_1 – irradiation time, sec; σ – cross section of radionuclide ^{60}Co, barn; t_o – irradiation time of monitor sec;

σ – activation cross section of reaction ^{59}Co (n, γ) ^{60}Co, barn;

λ – decay constant (^{60}Co); $T_{1/2}$ – half-life (^{60}Co), sec;

Note: $e^{\lambda t} = \frac{0,693}{T_{1/2}} t$

The most complete formula for determining the thermal neutron flux density is as follows:

$$F_{th} = \frac{A \cdot e^{\lambda \cdot t_{ex}} \cdot S}{0,6 \cdot \sigma \cdot R_\lambda \theta \cdot \varepsilon \cdot P \cdot m \cdot (1 - e^{-\lambda \cdot t_0}) \cdot \tau_m} - \frac{A \cdot e^{\lambda \cdot t_{\tilde{n}\tilde{e}d}} \cdot S_{Cd}}{0,6 \cdot \sigma \cdot R_\lambda \theta \cdot \varepsilon \cdot P \cdot m_{Cd} \cdot (1 - e^{-\lambda \cdot t_{0 \cdot Cd}}) \cdot \tau_{\tilde{n}\tilde{e}d}} \quad (2)$$

Where:

S - area of the detector photopeak without Cd screen, imp./sec;

S_{Cd} - area of detector photo peak in Cd screen, imp/sec;

A - atomic weight of Co-59;

λ - decay constant of the isotope Co-60, sec^{-1};

σ - Co-60 activation cross section, barn (1·10^{-24}cm²);

R_γ - gamma ray output;

θ - isotope content in the detector;

ε - analyzer sensor efficiency (3,53·10^{-3});

P - reactor power, MW;

m – detector mass, g(3·10^{-5});

m_{Cd} - mass of the detector in the Cd screen, g;

t_o - Detector irradiation time without Cd screen, sec;

$t_{o\,Cd}$ - detector irradiation time in the Cd screen, sec;

t_{ex} - detector exposure time without Cd screen, sec;

t_{exCd} - detector exposure time in the Cd screen, sec;

τ_m - measuring time of the detector without Cd screen, sec;

$\tau_{m\,Cd}$ - measurement time of the detector with Cd-screen, sec.

Methodology of radiochemical processing of irradiated targets and production of sulfur-35

To separate the target sulfur-35 radionuclide from radionuclide impurities, various radiochemical methods are used, such as precipitation, ion exchange, extraction, and sublimation.

The irradiated potassium chloride target was dissolved in 0.1 N hydrochloric acid solution and then adsorbed on a chromatographic column with aluminum oxide, m = 5.0 g. The chromatographic column was then washed with distilled water to remove K+ and Cl– ions. The ^{35}S radionuclide was eluted with 1 N ammonia solution. The eluate contained $(NH_4)_2{}^{35}SO_4$. Purification of the target radionuclide ^{35}S from the admixture of radionuclide P-32 is necessary. Radionuclide P-32 has a beta radiation energy of E_β = 1.71 MeV, which can greatly distort the results of measuring the activity of radionuclide S-35, because S-35 has a maximum energy of E_β = 0.167 MeV. Therefore, it is important to purify the target radionuclide from the impurity radionuclide P-32. It is known that for the extraction of radionuclide ^{35}S in the form of sulfate, a method is used based on the absorption of $^{35}SO_4$ ions on chromatographic aluminum oxide. A solution of irradiated NaCl in 0.5 N HCl is passed through a tube with Al_2O_3, on which both the target radionuclide ^{35}S and the impurity of radionuclide P-32 are sorbed. Then the column is washed with water, and then sulfur-35 is washed out with a 1.0 N ammonia solution. The P-32 radionuclide impurity remained sorbed on the sorbent. The sulfur yield is about 90%. The radiochemical purity of radionuclide S-35 reaches 99.9% [16]. The radionuclide ^{32}P can be obtained from the natural isotope sulfur-32 by neutron irradiation via the nuclear reaction ^{32}S (n, p) → ^{32}P [17], and in this context radionuclide ^{32}P is formed as a basic radionuclide

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

impurity to the target radionuclide ^{35}S, which is formed by a nuclear reaction ^{35}Cl (n, 2n) ^{34}Cl (Table 1).

The sodium chloride and magnesium chloride targets were processed similarly. The ampoule with irradiated carbon tetrachloride was opened after being cooled in liquid nitrogen (–195.75 °C), then transferred to a separatory funnel, and an aqueous solution of 1 N ammonia was added with vigorous stirring. Carbon tetrachloride was separated from the aqueous phase, which contained carrier-free S-35 in the form of $(NH_4)_2{}^{35}SO_4$. The aqueous phase was passed through a chromatographic column with aluminum oxide. The eluate contained sulfur-35, and the radionuclide impurity P-32 remained sorbed on the sorbent.

To measure the induced activity of the radionuclide ^{35}S, we used the beta-gamma spectrometer "Progress BG (II)" BDEB 3-2U with the software "Progress 5". The confidence limits of the total error of the measurement result of external beta radiation at a probability of 0.95 were within ±20%.

The formula for the definition activity of radionuclide S-35 is:

$$Q = \frac{0.6 \cdot F_t \cdot \sigma_t \cdot \theta \cdot m \cdot (1 - e^{-\frac{0.693 \cdot t_0}{T\frac{1}{2}}})}{A \cdot 3,7 \cdot 10^7} \quad (3)$$

where: Q -activity of S-35, mCi; F_t - thermal neutrons flux density, neutrons/cm²sec; t_o - irradiation time of sample, sec; $T_{1/2}$ - half-life S-35, sec; - enrichment of S-35, %; m - weight of target S-35, g; σ_t - activation cross section of reaction ^{35}Cl (n, p) ^{35}S, barn.

When calculating the induced activity of sulfur-35 using the given formula, there is no contribution of fast neutrons to the activation above the initial ingredient. In order to pay attention to the thermal neutrons on the activation of the irradiated target, we studied the cadmium ratio (R_{Cd}).

The measurement of the cadmium ratio R_{Cd} is found from the results of two measurements. First of all, a target without a cadmium sheath is placed in the neutron field. It is activated by both thermal and resonant neutrons. After determining the induced activity A_1 and holding for a period during which almost all active nuclei will decay, the target is wrapped in a cadmium sheath (screen) and placed back in the same place in the neutron field.

Cadmium has unusually high thermal neutron capture cross sections (7200 barns), and hence the radioactivity induced in cadmium-wrapped targets is approximately proportional to the intermediate neutron flux.

The cadmium ratio R_{Cd} is equal to the ratio of the effect of activation by thermal neutrons to the effect of activation by resonant neutrons:

To determine the cadmium ratio, 10 weighed portions of potassium chloride samples were irradiated with and without a cadmium screen under the same conditions: thermal neutron flux $F_n = 1\ 10^{13}$ neutrons/cm²sec and irradiation time t = 1 hour.

Knowing $R_{Cd,}$ it is easy to take into account the contribution of neutrons when irradiating a sample above the cadmium region of the neutron spectrum using the formula:

$$K_{Cd} = \frac{R_{Cd}}{Rcd - 1} \quad (4)$$

where: R_{Cd}- cadmium ratio, K_{Cd} -target activation coefficient over the cadmium region of the neutron spectrum (E > 1.0 eV).

The total effective activation cross-sectionσ_{eff} is formed by the product of the thermal neutron activation cross-section and the coefficient R_{Cd}.

The total effective activation cross-section δ_{eff}. It is formed by the product of the thermal neutron activation cross-section and the coefficient K_{Cd}. Therefore, the formula for calculating the induced activity of the radionuclide ^{35}S takes the following form:

$$Q = \frac{0.6 \cdot F_t \cdot \delta_t \cdot K_{Cd} \cdot \theta m \cdot \theta_1 \cdot (1 - e^{-\frac{0.693 \cdot t_0}{T\frac{1}{2}}})}{A 3,7 \cdot 10^7} \quad (5)$$

Where: Q - activity of S-35, mCi; F_t - thermal neutrons flux density, neutrons/cm²sec; t_o- irradiation time of sample, sec; $T_{1/2}$ - half-life S-35, sec; θ - enrichment of Cl-35, %; θ_1 - mass fraction of chlorine in a chlorine-containing compound; m - weight of target weight of chlorine-containing compound, g; δ_t - activation cross section of reaction ^{35}Cl (n, p) ^{35}S, barn.

Table 1: Nuclear reactions of formation of long-lived impurity radionuclides.

Radionuclide	$T_{1/2}$	Nuclear reaction
Cl-34	1.6 sec	^{35}Cl (n, p) ^{34}Cl
Cl-34	32.4 min	^{35}Cl (n, 2n) ^{34}Cl
Cl-34	37 min	^{37}Cl (n, 2n) ^{34}Cl
Cl-38	37 min	^{37}Cl (n, γ) ^{38}Cl
Cl-38	37 min	^{41}K (n, α) ^{38}Cl
P-32	14.5 days	^{35}Cl (n, 2n) ^{32}P
P-34	12 sec	^{37}Cl (n, α) ^{34}P
K-42	12.4 hour	^{41}K (n, γ) ^{42}K

Abbreviations and notes: Half life - $T_{1/2}$; (n, 2n) - a nuclear reaction in which a nucleus absorbs a neutron and emits two neutrons (this reaction usually occurs when nuclear material is irradiated with high-energy neutrons - fast neutrons); (n, p) - a nuclear reaction in which a nucleus captures a neutron (n) and emits a proton (p); a nuclear reaction (n, γ) in which a target nucleus captures a neutron and the excitation energy of the resulting nucleus is emitted as a γ-quantum; a nuclear reaction in which a nucleus captures a neutron (n) and emits an alpha particle (α).

Discussion

Figure 1 shows the energy spectrum of beta radiation of the radionuclide ^{35}S [18].

Figure 2 shows WWR-SM reactor core cartogramwhere indicated: 1 - 6-tube IRT-4M; 2 - 8-tube IRT-4M; 3 - 6-tube

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

IRT-4M with emergency protection rod; 4 - working element of the automatic control rod in a Be block; 5 - № 9 dry channel for irradiation; 6 - Be reflector block with plug (∅ = 44 mm); 7 - segmented Be reflector block with channel; 8 - Be reflector block withchannel (∅ = 60 mm); 9 - horizontal dry channel №3; 10 - beryllium reflector block with channel (∅ = 44 mm); 11 - lateral beryllium displacer; 12 - coolant (water).

Figure 3 shows a drawing of a 6-tube fuel assembly of the IRT-4M type and the placement of a special aluminum block container inside the IRT-4M. A characteristic feature of the special aluminum block container is the cooling of the ampoule with the irradiated carbon tetrachloride target by the running water of the first circuit, which enters the block container through the inlet and outlet openings.

Figure 4 shows the accumulation curve of sulfur-35 in a thermal neutron flux density of $0.8 \cdot 10^{14}$ n/cm²sec. As can be seen from Figure 4, the maximum specific activity of sulfur-35 of 3.312 Ci/g is achieved with irradiation for 2000 hours. Thus, by selecting several parameters, such as location in a standard channel (closer to the center of the active zone) or inside the fuel assembly in the reactor, sample packaging, irradiation time, and others, it is possible to achieve high results in obtaining high specific activity of sulfur-35.

Table 2 shows the main nuclear-physical characteristics of neutron activation detectors of thermal and resonance neutrons. For determining the thermal neutron flux density, the most convenient tracking monitors are the Co-59 and Au-197.

Table 3 shows nuclear-physical characteristics of the tracking monitor (detector) Co-59 with activated radionuclide Co-60. In experiments to determine the thermal neutron flux density, we used Co-59 monitors, which are stable at high temperatures and the most convenient with nuclear-physical characteristics that are convenient for calculations.

When irradiating targets of chlorine-containing compounds

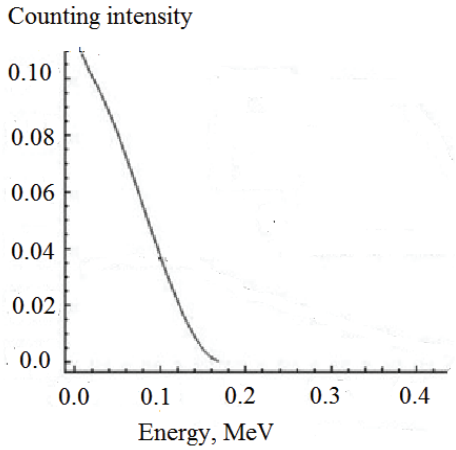

Figure 1: Beta radiation energy spectrum of the radionuclide ^{35}S [18]. From the energy spectrum of beta radiation of the radionuclide ^{35}S shown that it is evident that sulfur-35 has a maximum energy of E = 0.167 MeV.

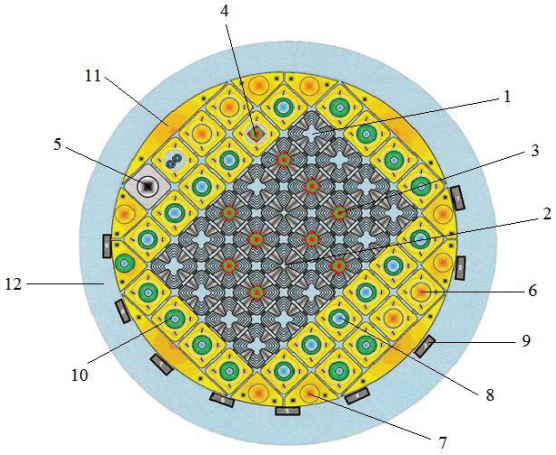

Figure 2: WWR-SM reactor core cartogram. 1 - 6-tube IRT-4M; 2 - 8-tube IRT-4M; 3 - 6-tube IRT-4M with emergency protection rod; 4 - working element of the automatic control rod in a Be block; 5 - № 9 dry channel for irradiation; 6 - Be reflector block with plug (∅ = 44 mm); 7 - segmented Be reflector block with channel; 8 - Be reflector block withchannel (∅ = 60 mm); 9 - horizontal dry channel №3; 10 - beryllium reflector block with channel (∅ = 44 mm); 11 - lateral beryllium displacer; ; 12 - coolant (water).

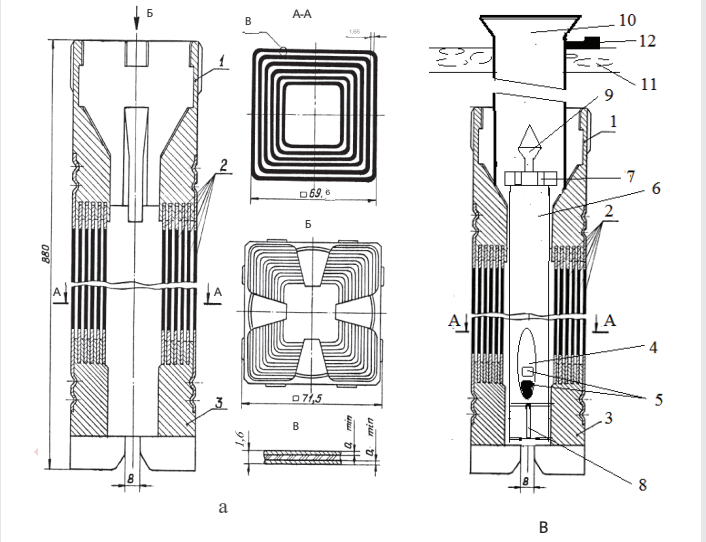

Figure 3: 3: 6-tube IRT-4M fuel assembly (a) and special aluminum block container (b).(A) - 6-pin fuel assembly type IRT-4M: 1-head; 2- fuel element; 3 – tail. (B) - special container for irradiation of carbon tetrachloride target: 1-head; 2- fuel element; 3 – tail; 4 -quartz ampulla; 5 - target of liquid carbon tetrachloride; 6 – special block container; 7 - coolant inlet hole; 8 – coolant outlet hole; 9 - container head; 10 - vertical guide pipe); 11 - reactor platform; 12 - bracket for fixing the guide pipe.

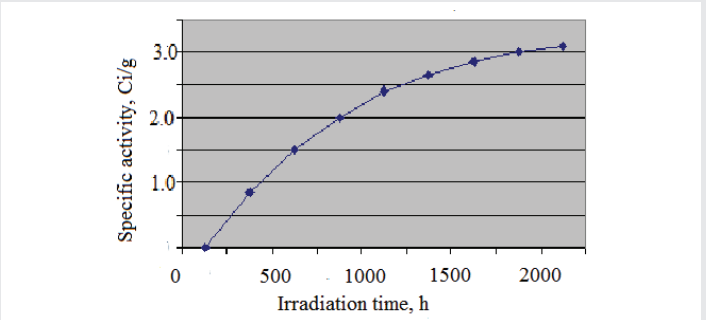

Figure 4: Accumulation curve of sulfur-35 in a thermal neutron flux density of $0.8 \cdot 10^{14}$ n/cm²sec. As can be seen in Figure 4, the maximum activity of the sulfur-35 radionuclide is achieved by irradiating a chlorine-containing target for 2000 hours.

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

with reactor neutrons, simultaneously with radionuclide ^{35}S, impurity radionuclides are formed from elementary elements included in the starting target, which are obtained by nuclear reactions (n, α), (n, n), (n, 2n). These nuclear states, when irradiating the initial target with fast neutrons with an activity of F_f > 10 MeV. However, the value of integral fast neutrons with energy F_f > 10 MeV in the neutron spectrum is 5-6 orders of magnitude less than that of thermal neutrons. Therefore, when irradiating chlorine-containing targets at the WWR-SM reactor, fast neutrons make the smallest contribution to the formation of radionuclides. In addition, when irradiating targets with neutrons, several radionuclides with a very short half-life are formed, which decay after 10 - 15 hours of exposure after irradiation. To obtain radionuclide ^{35}S with high specific activity and radiochemical purity, it is necessary to separate the whole product from the radionuclide impurities, especially from the radionuclide ^{32}P, the presence of which in the target product distorts the results of measuring the activity of radionuclide ^{35}S. Table 1 shows the nuclear reactions of the formation of long-lived impurity radionuclides.

Table 4 shows the cadmium ratio value for ^{35}S, where you can see that the cadmium ratio for sulfur-35 is average - 9.37, i.e., the nuclear reaction ^{35}Cl (n, p) ^{35}S occurs mostly by thermal neutrons with energy En 0.025 eV.

Table 5 shows the values of the induced activity and the practical output activity of sulfur-35 depending on the irradiation time in a thermal neutron flux of $1 \cdot 10^{14}$ n/cm²·s. As can be seen from the table, the highest specific activity of the sulfur-35 radionuclide occurs when carbon tetrachloride targets are irradiated.

The simplicity of the extraction of sulfur-35 from the irradiated target is because the radiochemical extraction process is carried out in a protective box with lead gloves by hand: irradiated liquid carbon tetrachloride is poured into a glass separatory funnel, into which distilled water is also

Table 2: Main nuclear-physical characteristics of neutron activation detectors of thermal and resonance neutrons.

Number in order	Nuclear reaction	σthermal, barn	T1/2	E_γ, keV (k_γ, %)
1	$^{176}Lu(n,\gamma)^{177}Lu$	2100	6,71days	113(6,60),208(11,0)
2	$^{151}Eu(n,\gamma)^{152}Eu$	3211	9,3h	842(14,6)
3	$^{197}Au(n,\gamma)^{198}Au$	98,7	2,696days	412(95,5)
4	$^{152}Sm(n,\gamma)^{153}Sm$	206	47,1h	103(28,2)
5	$^{186}W(n,\gamma)^{187}W$	37,8	23,9h	686(29,7)
6	$^{139}La(n,\gamma)^{140}La$	8,95	40,26h	1596(95,3)
7	$^{59}Co(n,\gamma)^{59}Co$	37,2	5,272year	1133(100),1173(100)
8	$^{63}Cu(n,\gamma)^{64}Cu$	4,51	12,7h	511(36,8)
9	$^{23}Na(n,\gamma)^{24}Na$	0,527	14,96h	1368(100),2754 (99)
10	$^{50}Cr(n,\gamma)^{51}Cr$	15,9	27,7days	320(9,83)
11	$^{45}Sc(n,\gamma)^{46}Sc$	27,3	84,0days	889(100),1121(100)

Abbreviations and notes: σthermal- activation cross section for thermal neutrons; E_γ-gamma quanta energy; A nuclear reaction is a process of interaction of an atomic nucleus with another nucleus or elementary particle, which may be accompanied by a change in the composition and structure of the nucleus.

Table 3: Nuclear-physical characteristics of tracking monitor Co-59 with activated radionuclide ^{60}Co.

Nuclear-physical characteristics	Meaning
Isotopic concentration Co-59	100 %
Nuclear reaction	$^{59}Co(n,\gamma)^{60}Co$
Activation cross section, b	0.35 - 0.575
Radionuclide ^{60}Codecay constant	$4.169 \cdot 10^{-9} sec^{-1}$
Radionuclide ^{60}Co half-life, $T_{1/2}$	46182.43 hour
Output of gamma-quantum, R_γ	0.998
Energy of gamma-quantum, E_γ	1332 keV
Thermal neutrons' energy, E_n	≤0.025 eV
Endurance time of irradiated target, hours	48÷72

Abbreviations and notes: b – barn. 1 barn is equal to 10^{-28} m² = 10^{-24} cm² = 100 fm² (approximate size of an atomic nucleus); eV – electronvolt; keV - kiloelectronvolts ($1 \cdot 10^3$ eV).1.0 eV is equal to the energy required to transfer an elementary charge in an electrostatic field between points with a potential difference of 1 V.

Table 4: Cadmium ratio value for radionuclide^{35}S.

Experiment number	Activity of ^{35}S, Bk/g		
	In the cadmium screen	Without a cadmium screen	R_{Cd}
1	3.830	37.0	9.53
2	4.0	45.484	11.37
3	3.740	30.850	5.25
			Average value R_{Cd} =9.37

Abbreviations and notes: Bq: Becquerel. Bq is a unit of radiation measurement in the International System of Units (SI). It corresponds to one radioactive decay per second; R_{Cd}:Cadmium ratio;

Table 5: The value of the induced activity and the practical output activity of sulfur-35 depending on the irradiation time under a thermal neutron fluxof $1 \cdot 10^{14}$neutrons/cm^2 ·sec.

NO	Irradiation time, h	Accumulation factor	Activity of chlorine-containing compound, Ci/g		Practical activity output, Ci/g	
			CCl_4	KCl	CCl_4	KCl
1	100	0.0327	0.222	0.114	0.275	0.125
2	200	0.0644	0.438	0.225	0.410	0.215
3	300	0.0909	0.619	0.318	0.733	0.411
4	400	0.124	0.845	0.434	0.905	0.388
5	500	0.153	1.042	0.536	1.012	0.573
6	600	0.181	1.233	0.634	1.80	0.720
7	700	0.207	1.410	0.725	1.420	0.745
8	800	0.233	1.587	0.817	1.550	0.730
9	900	0.259	1.765	0.907	1.818	1.05
10	1000	0.283	1.928	0.991	1.960	1.200
11	1200	0.329	2.242	1.153	2.125	0.950
12	1400	0.372	2.535	1.303	2.421	1.216
13	1600	0.412	2.807	1.703	2.855	1.621
14	1800	0.450	3.066	1.577	3.087	1.610
15	2000	0.468	3.312	1.713	3.422	1.645

Abbreviations: h: hour; Ci/g: Curie of radionuclide ^{35}S per 1 gram irradiated target.

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004

poured. Next, shake the separatory funnel several times. Carrier-free radionuclide ^{35}S passes from carbon tetrachloride into the aqueous phase, the aqueous phase is easily separated from the organic phase (by draining the radioactive water), and then the activity of sulfur-35 is determined on a gamma-beta spectrometer. During radiochemical processing of the target by the extraction method, the organic phase does not mix with the aqueous phase, and the coefficient of extraction of the radionuclide ^{35}S from the organic phase to the aqueous phase is high:

$$K_{ext} = \frac{1000\,mCi}{80\,mCi} = 12.5 \quad (6)$$

The recovery factor of sulfur-35 from the organic phase to the aqueous phase is 12.5, and the target carrier-free sulfur-35 radionuclide has a yield of > 92%.

Conclusion

Experimental data show that the yield of sulfur-35 activity per 1 gram of chlorine-containing compound is the highest when irradiating a carbon tetrachloride target with neutrons, since the mass fraction of sulfur in Cl_4 is the highest - 92.5%, while in $MgCl_2$ it is 74%, in NaCl it is 60% and in KCl it is 45%. When irradiating a target made of KCl, NaCl, or $MgCl_2$ with neutrons, a chromatographic column with a sorbent is required to isolate sulfur-35 and purify it from radionuclide impurities, the use of which reduces the yield of the target product S-35 to 60% - 70%. And in the case of using a carbon tetrachloride target, the extraction of the target radionuclide ^{35}S from the irradiated target is carried out by a simple water extraction method, which does not require complex radiochemical operations. However, carbon tetrachloride target samples must be irradiated in vertical channels of reactor with mandatory cooling by water of the reactor's primary circuit, since at high temperatures carbon tetrachloride passes from the liquid phase to the gaseous phase (the boiling point of CCl_4 is 76.6 °C), which was observed by us during the irradiation of targets in the "dry" channels of the WWR-SM reactor.

Thus, the highest yield of sulfur-35 activity per 1.0 g of chlorine-containing compound 3.312 Ci/g is achieved by irradiating CCl_4 targets under the following conditions: thermal neutron flux density is $\geq 1 \cdot 10^{14}$ n/cm^{2}sec, irradiation time is ~2000 h, nominal reactor power is 10 MW, and irradiation of a quartz ampoule with the target in a vertical reactor channel with mandatory cooling of the target with running first loop water. The isolation of sulfur-35 without a carrier from irradiated carbon tetrachloride was carried out using the water extraction method, which is the simplest and does not require complex technological operations. This radiation technology is used to produce highly active sulfur-35 radionuclide without a carrier in research reactors.

Acknowledgment

The authors thank senior research fellows Abdusalyamov N. and Ashrapov T.B. for their assistance in consulting during the experiments.

References

1. Schubert M, Kneller K, Tegen I, Terzi L. Variability of cosmogenic 35S in rain—implications for using radioactive sulfur as a natural indicator of groundwater residence time. Water. 2020;12(10):2953. Available from: https://www.mdpi.com/2073-4441/12/10/2953

2. Collins C, Cunningham N. Modelling the fate of sulphur-35 in crops. 1. Calibration data. Environ Pollut. 2005;133(3):431–7. Available from: https://doi.org/10.1016/j.envpol.2004.07.001

3. Hershey A, Chase M. Independent functions of viral protein and nucleic acid in growth of bacteriophage. J Gen Physiol. 1952;36(1):39–56. Available from: https://doi.org/10.1085/jgp.36.1.39

4. Wittwer AJ, Stadtman TC. Biosynthesis of 5-methylaminomethyl-2-selenouridine, a naturally occurring nucleoside in Escherichia coli tRNA. Arch Biochem Biophys. 1986;248(2):540–50. Available from: https://doi.org/10.1016/0003-9861(86)90507-2

5. Levin VI. Obtaining radioactive isotopes. Moscow: Atomizdat. 1972;130.

6. Gledenov YuM, Salatsky VI, Sedyshev PV, Shalansky IP. Measurement of the reaction cross section 35Cl(n,p)35S for thermal neutrons. 1996. Available from: https://inis.iaea.org/records/0jybg-6bc08

7. Oh MA, Prelas JB, Rothenberger ED, Lukosi JJ, Montenegro DE, et al. Theoretical maximum efficiencies of optimized slab and spherical betavoltaic systems utilizing sulfur-35, strontium-90, and yttrium-90. Nucl Technol. 2012;179:9.

8. Van den Berghe S, Leenars A, Koonen E, Sannen L. From high to low enriched uranium fuel in research reactors. Adv Sci Technol. 2010;73:78–90. Available from: http://dx.doi.org/10.4028/www.scientific.net/AST.73.78

9. Yuldashev BS, Salikhbaev US, Baytelesov SA, Dosimbaev AA, Khalikov UA. Account and control nuclear materials at the WWR-SM reactor in the Institute of Nuclear Physics, Tashkent. In: Safety Related Issues of Spent Nuclear Fuel Storage: Proceedings of the NATO Advanced Research Workshop. Almaty, Kazakhstan; 2005 Sep 26–29;143–5. Available from: https://link.springer.com/chapter/10.1007/978-1-4020-5903-2_9

10. GOST 4234-77. Reagents. Potassium chloride. Specifications. Moscow: Standards Publishing House. 2001;28.

11. GOST 4233-77. Reagents. Sodium chloride. Specifications. Available from: https://runorm.com/catalog/1004/736307/

12. GOST 4209-77. Reagents: Magnesium chloride 6-aqueous. Specification. Moscow: Standards Publishing House. 1977;24.

13. GOST 20288-74. Reagents. Carbon tetrachloride. Specifications. Moscow: Standards Publishing House. 1987;17.

14. Yuldashev BS, Ashrapov TB, Mirzaev NM, Ashrapov UT. Preliminary Patent Republic of Uzbekistan. No. IDP/04633. 2000.

15. Aliev AI, Drapkin VI. Handbook of nuclear data for neutron activation analysis. Washington (DC): I.P.S.T.; 1970;174. Available from: https://www.osti.gov/biblio/4660344

16. Skoblov YS. Radioactive isotopes in physical and chemical biology. Zbio. 2017. Available from: http://zbio.net/bio/001/004.html#a8

17. Levin VI. Obtaining radioactive isotopes. Moscow: Atomizdat. 1972;132.

18. Silaev ME, Chertkov YuB. Devices and methods of physical measurements. Tomsk: Publishing House of Tomsk Polytechnic University. 2008.

Citation: Ashrapov UT, Malikov SR, Mirzaev BB. Obtaining Sulfur-35 without a Carrier from Chlorine-containing Chemical Compounds. Insights Nucl Energy Sci Eng. 2025;3(1):001-007. Available from: https://dx.doi.org/10.17352/inese.000004