A half-century of radioisotope neutron sources in China

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Abstract: Near 50 years history of the development of radioisotope neutron sources in China is briefly reviewed. The structure design, preparing technology and production status of routine neutron sources including ^{210}Po-Be sources, ^{210}Po mock fission sources, ^{241}Am-Be sources, ^{238}Pu-Be sources, ^{252}Cf spontaneous fission sources and other special-shape neutron sources are summarized. In addition, the prospects of development on radioisotope neutron source in China are predicted from the needs of nuclear power construction, oil well-logging, neutron moisture gauge and neutron brachytherapy.

Key words: radioisotope neutron source; (α, n) source; spontaneous fission source; source core preparing; source capsule sealing; quality control; oil well-logging; neutron moisture gauge; neutron brachytherapy

1 Introduction

The mystery of nuclear structure was revealed by the discovery of neutrons in 1932. The first observed neutrons by Chadwick were produced through the bombardment of beryllium target with polonium-210 alpha particles. The nuclear reaction is as follows:

$$^{9}\mathrm{Be} + {}^{4}\mathrm{He} \rightarrow {}^{12}\mathrm{C} + \mathrm{n} + 5.71\ \mathrm{MeV}$$

It is a typical method for the preparation of (α, n) neutron source. The most common target is beryllium. In the 40's - 50's of the 20th century, radioisotope neutron sources were manufactured by naturally occurring alpha emitters, such as ^{210}Po, ^{226}Ra and its decay products. Along with the discovery of transuranium elements, the available alpha emitters were increased. The initial use of ^{226}Ra-Be and ^{210}Po-Be sources were gradually substituted by ^{239}Pu-Be, ^{241}Am-Be, ^{242}Cm-Be, ^{244}Cm-Be and ^{238}Pu-Be source after 1960's. Subsequently, the heavy nuclide, ^{252}Cf became widely available through its production in high flux nuclear reactors in 1970's. The advantages of ^{252}Cf spontaneous fission sources are its high neutron output and small physical size. Besides (α, n) sources and spontaneous fission sources, the third kind of neutron source is called (γ, n) source, or photoneutron source. Up to now, most of them are not used except ^{124}Sb-Be source[1,2]. Comparing with the reactor and accelerator neutron sources, the advantages of radioisotope neutron source are of small size, quite portable, reliable neutron yield and relatively low in cost. So they have been widely used in industry, agriculture, medicine and scientific research for many years.

The research of polonium started in China early in the 60's of the 20th century. In 1958, 10 MW heavy-water reactor reached the critical. It offers a good condition for the preparation of radioisotopes including polonium. After solving the chemical separation techniques of polonium from irradiated Bi_2O_3 targets, ^{210}Po α sources, ^{210}Po-Be neutron sources and ^{210}Po mock fission sources were fabricated respectively in the middle of 1960's. The "volatilization technology" was carried into effect for the preparation of ^{210}Po-Be sources, because of the breakthrough for the sealing technique of polonium source. ^{210}Po-Be sources had been produced in batches since 1967. It met the urgent needs of exploration of oilfield and mineral resource.

Depending on the development of well logging in the world and the requirement of domestic users, the long-lived ^{241}Am-Be neutron source began to study in the mid of 1970's. The special production line was set up and the production capability of ^{241}Am-Be sources had been formed. ^{210}Po-Li and ^{210}Po-LiF neutron sources were developed and the preparing technology of ^{210}Po mock fission source was improved by the end of 1970's. A pair of ^{210}Po-Be started up source rods were manufactured and PWR start-up of the first nuclear power station in China was accomplished successfully at the beginning of 1990's. In order to adapt to the fast development of nuclear power and nuclear medicine, the raw materials and preparing techniques of ^{252}Cf neutron source were introduced to China through the mode of cooperation between China and Russia. Initial batch ^{238}Pu-Be neutron sources were produced with "dry method" technology in 1979. The "wet method" technology was tested again in 2006. Up till

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now, all normal radioisotope neutron sources can be manufactured in China. The variety and amount of products continue to increase. Fig. 1 shows the different kinds of radioisotope neutron sources (specimens) made in China.

The fabricating techniques of radioisotope neutron sources including structure design, source core preparation, sealing technology and quality control are entirely summarized. The variety of products and present status of production are simply introduced. The prospects of radioisotope neutron sources in China are predicted from the investigation of different applications.

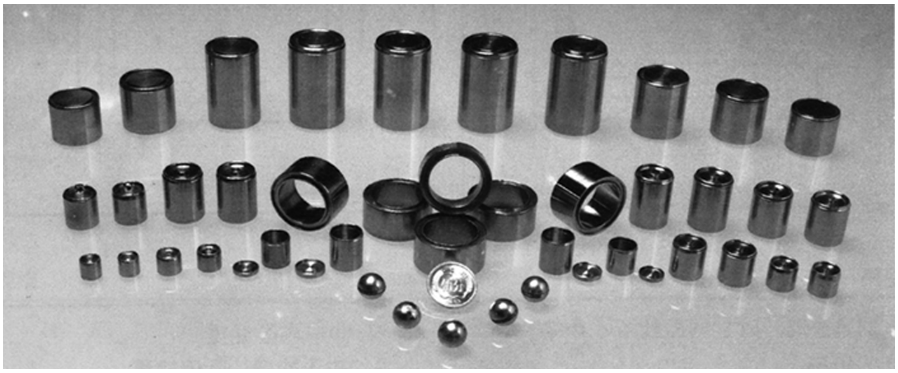


Fig. 1 Different kinds of radioisotope neutron sources (specimens) made in China

2 Natural radioisotope (α, n) neutron sources

Initial (α, n) neutron sources were prepared abroad with ^{210}Po and ^{226}Ra from naturally occurring radioisotopes. Among them, ^{226}Ra-Be neutron sources were utilized in many fields. Beryllium is common target, because it has high neutron output by bombarding of alpha emitters. The drawbacks of ^{226}Ra-Be sources were its stronger gamma emission, higher price and harmful radon gas leakage, so it was superseded by ^{210}Po-Be sources which had the advantages of higher output, small size and lower gamma emission. Especially, ^{210}Po can be produced by artificial method[3,4]. The drawback of ^{210}Po is shorter half-life (138.4 d). Depending on the practical situation of China, ^{210}Po-Be neutron sources and ^{210}Po mock fission sources were studied early in 60's of the 20th century. Then, ^{210}Po-Li and ^{210}Po-LiF neutron sources were trial-manufactured late in 1970's.

2.1 ^{210}Po-Be neutron sources

It is imperative that the fabrication of ^{210}Po-Be neutron sources should have polonium-210 materials. The extraction of ^{210}Po from irradiated Bi_2O_3 targets was selected in China. A lot of chemical separation techniques between polonium and bismuth were studied, such as coprecipitation, solvent extraction, TiO_2 chromatography, volatilization, spontaneous deposition, and electrodeposition[5-8]. Among them, the spontaneous plating with bismuth powder had been used in chemical separation process of polonium-210, because of its convenient operation, high extracted rate, and excellent separating effect. In addition, α radiation did not influence for the operation[9,10]. As regards electrodeposition method, it had been applied to the purification and concentration of polonium-210 as well as the manufacture of ^{210}Po α sources[11]. Early work site of extracting ^{210}Po from irradiated Bi_2O_3 targets is shown in Fig. 2.

The "wet method" and "sandwich method" were used in the fabrication of ^{210}Po-Be neutron sources early in the 1960's[12]. For the wet method, it was based on the electrochemical replacement reaction between beryllium metal and $^{210}Po^{4+}$, the polonium-210 was spontaneously deposited on beryllium powder in diluted nitric acid solution. However, tardily dissolved phenomena of beryllium powder were observed. It influenced the deposition of ^{210}Po. In order to decrease contact time between $^{210}Po^{4+}$ and beryllium powder, "beryllium column" spontaneously deposited method was adopted. After all, this technology had not been used in the practical production of ^{210}Po-Be neutron sources, because the operator dose was too high and the weld of inner capsule was easily contaminated in loading[13].

For the "sandwich method", ^{210}Po plating foil (Pt or Au backing) was inserted into beryllium powder and then sealed in double capsules. The neutron yield was lower because the components were poorly mixed. In order to increase the neutron yields, it was necessary to further volatilize polonium-210 on beryllium powder. Therefore, sealing of neutron source became a key technique. The soft soldering, cold (pressing) welding, brazing and decomposition of nickel carbonyl [$Ni(CO)_4$] had been tested (copper as the material of capsule). Most of them could not withstand severe tests of high temperature, high pressure and corrosion-resistant etc. Subsequently, tungsten-inert-gas (TIG) welding or called argon-arc welding was developed in 1966[14]. The materials of capsule were stainless steel (1Cr18Ni9Ti). An argon arc welding machine (scale of current regulation: 1 ~ 50 A) was fabricated. Passing through a series of experiments including the comparison of weld joint type, design of welding devices (Fig. 3) and selection of welding parameters, closure of polonium sources was carried out satisfactorily.

Fig. 2 Early work site of extracting ^{210}Po from irradiated Bi_2O_3 targets

Fig. 3 Argon arc welding device

"Volatilization method" had been applied to the preparation of ^{210}Po-Be neutron sources in batches since 1967. This process included ^{210}Po materials extraction, ^{210}Po α sources preparation, source capsule encapsulation, high temperature volatilization and quality administration. At first, the irradiated Bi_2O_3 targets were dissolved in concentrated hydrochloric acid, then added bismuth powder (80 ~ 100 mesh) to the solution after denitration by formic acid. The deposited efficiency of polonium is about 90 % under mechanically stirring condition. Later, the extraction technique of ^{210}Po was improved. The raffinate of stirring spontaneous deposition was passed through "bismuth column", the average deposition efficiency of polonium was increased to (98.6 ± 0.05) %[15]. In order to decrease bismuth content of polonium dissolved solution from "primary spontaneous deposition", "secondary spontaneous deposition" was needed. In general, the small column (loading ~1 g bismuth powder) was used. Bismuth powder contained ^{210}Po was dissolved in 9 mol/L HNO_3. Then, ^{210}Po was deposited on cathode (Au or Pt foil) by means of the electrolysis method (controlling cathode potential) or "internal electrolysis" method[16]. Obtained ^{210}Po α source was wrapped into aluminum foil and inserted to inner capsule loaded beryllium powder. Closure was conducted with TIG welding. The procedure for outer capsule welding was a duplication of the inner capsule welding. In order to ensure the quality of sealing, decontamination and leakage were tested after each welding step. Subsequently, ^{210}Po-Be neutron source was heated to 700 ~ 800 ℃ to volatilize polonium-210 on beryllium powder after being put into a high temperature furnace and filled with helium in it. Neutron emissions were monitored by BF_3 counter during the heating process.

Theoretical neutron yield of ^{210}Po-Be source is 7.7×10^4 n/s· GBq (2.85×10^6 n/s· Ci, 1 Ci = 3.7×10^{10} Bq). The practical yield is up to 6.75×10^4 n/s· GBq. (2.5×10^6 n/s· Ci). There are four types (nominal dimension: Φ 10 mm × 10 mm, Φ14 mm × 14 mm, Φ 16 mm × 19 mm, Φ 20 mm × 28 mm) and seven varieties of products. Neutron emission is from 4×10^4 n/s to 2.0×10^7 n/s.

2.2 ^{210}Po mock (^{235}U) fission sources

A mock fission neutron source is required for some physical tests and instrument calibration. For the fission spectrum of uranium-235, the average neutron energy is 1.867 ± 0.015 MeV and peak value is 0.8 MeV. The (α, n) neutron sources combined ^{210}Po or other α emitter with light element targets such as Li, Be, B, F, etc., have different neutron spectrums. If above mentioned light elements were mixed with ascertain proportion, the ^{210}Po mock fission spectrum could be obtained. Among them, main contributions of lower and middle neutron energy parts are from Li and F, while more then 3 MeV neutron energy parts are from B and Be.

There are both "wet method" and "dry method" for the preparation of ^{210}Po mock fission sources. The wet method (evaporation method) had been used when sealing problem of neutron sources at high temperature was not yet solved. The prescription of $NaBF_4$ and Na_2BeF_4 were selected as mixed targets at beginning. Later, the prescription of mixed targets including $NaBF_4$, LiF and Be were adopted and the requirements of neutron spectrum were satisfied[16]. Evaporation process was carried on in a platinum crucible. At first, ^{210}Po plating foil (Au backing) was dissolved in hydrofluoric acid. Next, mixed targets (~0.6 g) were added to acid solution and heated to 90 ~ 100 ℃ for 60 min in a vacuum system. Then continue to heating at 120 ℃ for several minutes. The cap was opened after cooling and Pt crucible was taken out. The powders were loaded into copper capsule and sealed by cold (pressure) welding after grinding[12]. However, longer operation time and too much lose of polonium were drawbacks of wet method.

By the end of 70's, the wet method was substituted by the dry method (high temperature volatile method)[17]. The prescription of targets containing LiF, B and Be were tested. These targets can withstand high temperature. Polonium plating foil (Au backing) was inserted into mixed targets and doubly encapsulated in welded stainless steel capsules by TIG. Subsequently, heating at 700 ℃ in high temperature furnace, polonium was volatilized to targets. The neutron spectrum of ^{210}Po mock fission source prepared by dry method was similar to one of ^{235}U and ^{239}Pu. Also, neutron spectrum of dry method was the same to the one of wet method. Obviously, simple technology, stable neutron emission and small operation dose are the advantages of dry method.

The products of ^{210}Po mock fission neutron source has three nominal dimensions (Φ10 mm × 10 mm, Φ 14 mm × 14 mm, Φ16 mm × 19 mm). Average neutron energy is 1.7 ~ 1.9 MeV and peak value was 0.9 ~ 1.1 MeV. Neutron emission was up to 1.0×10^6 n/s, neutron yield > 1.2×10^5 n/s· Ci.

2.3 ^{210}Po-Li and ^{210}Po-LiF neutron sources

In order to meet the needs of lower neutron energy for some physical tests and neutron instrument calibration, ^{210}Po-Li and ^{210}Po-LiF neutron source were developed in the end of 1970's[18]. Volatile method was adopted. At first, polonium plating foil (Pt backing) was inserted into target material (Li metal or granular LiF). Then, it was doubly encapsulated in argon arc welded stainless steel capsules. Moreover, the source was placed into high temperature furnace and heated to ~700 ℃. Finally, polonium was distributed uniformly to the targets. According to the international and national standard of radioactive sources, leakage test and contamination monitor were carried out before and after heating for each neutron source.

The nominal dimension of both neutron sources is Φ 16 mm × 19 mm. Neutron spectrum was measured by proportional counter (containing hydrogen). Average neutron energy of ^{210}Po-Li and ^{210}Po-LiF neutron sources are 0.378 MeV and 0.585 MeV respectively. The neutron emission of ^{210}Po-Li source is 2.0×10^5 n /s and the neutron emission of ^{210}Po-LiF source is 1.0×10^6 n/s.

3 Artificial radioisotope (α, n) neutron sources

Isotopes of transuranium such as ^{239}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm, and ^{238}Pu[19-22] can be produced by artificial method. All of them belong to high energy α emitters and they can be used to fabricate (α, n) neutron sources. Comparing with strong γ radiation ^{226}Ra, lower specific activity ^{239}Pu and short half-life ^{210}Po (138.4 d), the characteristics of ^{241}Am α emitter are much better[23,24]. The advantages of ^{241}Am-Be neutron source are of its lower γ radiation, longer half-life (433 a) and stable neutron output. The calibration of decay is also simple. However, the bigger physical size is its drawback, because the specific activity of ^{241}Am is a little bit lower (3.43 Ci/g). It is no problem for use of oil well logging and industrial process control. The drawbacks of ^{241}Am-Be neutron source can be just overcome by ^{238}Pu-Be neutron source. The merits of plutonium-238 are its longer half-life (87.7 a) and higher specific activity (17.4 Ci/g)[25-27]. In the 1970's, the powder metallurgical technology of fabricating ^{241}Am-Be neutron source was studied in detail. ^{238}Pu-Be neutron source was developed again using "wet method" technology in new century.

3.1 ^{241}Am-Be neutron sources

Initial ^{241}Am-Be neutron sources were fabricated by "wet method" in the world. Afterward, this method was substituted by "dry method". Currently there are three kinds of fabricating techniques, that is pressed loading, alloy and ceramic form[28-33].

"Pressed pellet" technology had been used for 30 years in China[34]. The mixture of $\mathrm{AmO_2}$ and beryllium metal powder was not directly pressed to inner capsule, but rather pressed into disc (cylindrical pellet). Comparing with the "pressed loading", the advantages of pressed pellet technology are more convenient, easier to loading and less contamination of weld joint. In addition, the numbers of pellet can be increased or decreased depending on the requirement of neutron emission. The neutron yield of pressed pellet technology is higher than alloy and ceramic technology.

The flow sheet of ^{241}Am-Be neutron sources production is shown in Fig. 4. Firstly, $^{241}\mathrm{AmO_2}$ and beryllium metal powder were mixed intimately in an appropriate proportions at "Y" type mixer (rotating 20 ~ 30 min by motor). Then, the mixture was pressed into a pellet by single floating die. Depending on the requirement of source intensity, the pellets were loaded into stainless steel capsule and encapsulated with three layer capsules. Finally, severe quality control was carried out.

The influence of $^{241}\mathrm{AmO_2}$ materials on neutron yield was observed. At beginning, the neutron yield of two neutron sources were less then 2.0×10^6 n/s• Ci. Passing through the analysis, more than 100 mesh $^{241}\mathrm{AmO_2}$ granules were found in the ^{241}Am materials. Therefore, grinding step was added to the process, neutron yield was increased to $(2.0 \sim 2.5) \times 10^6$ n/s• Ci.

The influence of Be/$^{241}\mathrm{AmO_2}$ (mass ratio) on neutron yield was tested. When a mass ratio of Be / $^{241}\mathrm{AmO_2} > 10$, the tendency of neutron emission increasing started to slow up. In order to consider both neutron yield and volume of source, a ratio of Be/$^{241}\mathrm{AmO_2}$ mass in excess of 10 ~ 12 was suitable.

The influence of compressive strength on the quality of core pellet was also tested. When pressure > 1.1 t/cm^2, mixture of $^{241}\mathrm{AmO_2}$ and beryllium metal powder started to form pellet, but it is easy to split in the out of die. When pressure > 1.5 t/cm^2, pellet has certain strength and it is not easy to split. When pressure > 2.0 t/cm^2, pellet not only has higher strength, but has metal luster on the surface. In the practical production, the pressure of 4.5 ~ 6.0 t/cm^2 was selected, the density of pellets is up to 1.4 ~ 1.5 g/cm^3.

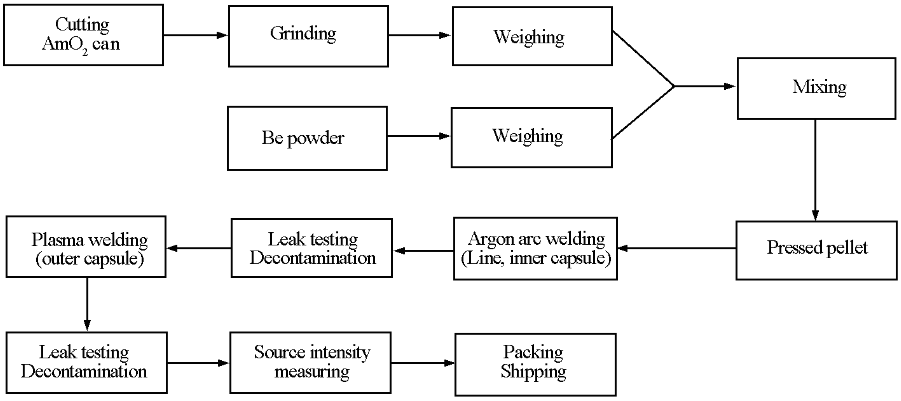


Fig. 4 Flow sheet of ^{241}Am-Be neutron sources

The encapsulation of three layer stainless steel capsules (1Cr18Ni9Ti) is adopted for ^{241}Am-Be neutron sources, because of its long-lived. Lining and inner capsule are sealed by TIG welding. Outer capsule is sealed by the plasma arc welding (Fig. 5)[35]. The advantages of plasma arc welding are of high temperature gradient, large energy density, stable arc column and faster welding speed. It can get excellent weld with bigger melted depth, narrow heating influence area and higher weld joint strength. In the process of welding, the mixed gas ($\mathrm{Ar} + 0.5\%\ \mathrm{H_2}$) was used in "ion gas" and "protective gas", it can prevent melting pool from oxidation, improve apparent quality of weld and raise the welding speed.

Neutron spectrum of ^{241}Am-Be neutron source was measured by recoil proton scintillation spectrometer. Average neutron energy is 4.35 MeV. The safety performance test of prototype source was up to GB/E56535[34-36]. In addition, special test combined temperature (200 ℃) with external pressure (2×10^2 MPa) was conducted. The practical test of ^{241}Am-Be sources in the first super-deep well (7 175 m) of

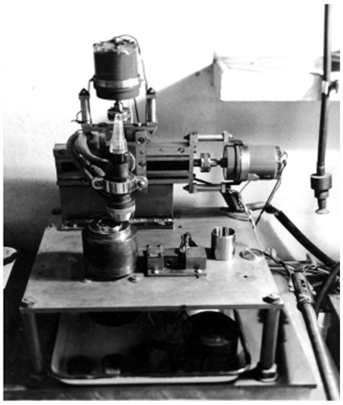


Fig. 5 Plasma arc welding device

China was passed.

^{241}Am-Be neutron sources had been produced in batches since 1978. New production line of ^{241}Am-Be sources was founded in 1990 (Fig. 6). A series of products had been formed. At present, there are 6 types and more than 20 varieties of products. The maximum activity of ^{241}Am is 740 GBq (20 Ci) and its nominal dimensions are Φ 26.5 mm × 111 mm or Φ 43 mm × 169.5 mm respectively.

Fig. 6 Production line of ^{241}Am-Be neutron source

3.2 ^{238}Pu-Be neutron sources

Utilizing $^{238}\mathrm{PuO_2}$ materials (~12 Ci/g) made in China, ^{238}Pu-Be neutron sources were trial-fabricated in the middle of 1970's. "Pressed pellet" technology of ^{241}Am-Be neutron source was adopted. The mixture of $^{238}\mathrm{PuO_2}$ and beryllium metal powder was pressed into pellets and then doubly encapsulated in stainless steel capsules welded by TIG welding. Average neutron yield was up to 1.83×10^6 n/s·Ci for 16 ^{238}Pu-Be neutron sources.

HTA Company utilized imported $^{238}\mathrm{PuO_2}$ materials to fabricate ^{238}Pu-Be neutron sources by "dry method" in 2006. They observed that the neutron yield was lower. Therefore, the "wet method" technology was tested. At first, $^{238}\mathrm{PuO_2}$ materials were dissolved in 7 mol/L $\mathrm{HNO_3}$ (forming a deep-green color solution), but it is hard to dissolve $^{238}\mathrm{PuO_2}$. Later, this step was substituted by catalytic electrolysis and the dissolved time can be shortened from 15 d to 3 ~ 5 h. The beryllium metal powder was put into plutonium nitrate solution. In order to decrease the surface tension, alcohol was added at the same time. Plutonium was precipitated with ammonia water and then dried. The pellets were made by pressing and calcined at 500 ~ 550 ℃. Finally, pellets were sealed in double stainless steel capsules welded by TIG. At present, the improvement of "wet method" technology is still carried on.

More than 20 ^{238}Pu-Be neutron sources were produced with wet method. Among them, 15 ^{238}Pu-Be neutron sources had been used in oil well logging. The maximum activity is 740 GBq (20 Ci) and its nominal dimensions are the same to the ^{241}Am-Be sources. Other nominal dimensions are Φ 16 mm × 19 mm (for ~1.0×10^6 n/s ^{238}Pu-Be sources) and Φ 30 mm × 30 mm (for 1.0×10^7 n/s ^{238}Pu-Be sources).

4 ^{252}Cf spontaneous fission sources

^{252}Cf neutron source relies on its self fission to produce neutrons. The neutron spectrum of ^{252}Cf is similar to the fission spectrum of ^{235}U. Average neutron energy of ^{252}Cf source is about 2.348 MeV. The effective half life of ^{252}Cf is 2.65 a including α decay (96.9 %) and spontaneous fission (3.1 %). Its specific neutron yield is very high (2.35×10^{12} n/s·g). In addition, not any targets are needed, so that ^{252}Cf can be made as a neutron source which has the highest neutron emission and the smallest physical size among all radioisotope neutron sources[37,38].

Both China Institute of Atomic Energy and Research Institute of Atomic Reactors of Russia (CIAE-RIAR) took the mode of joined venture, the Beijing CIAE-RIAR Radioisotope Technology Co., Ltd. was established in 1992. RIAR side provided ^{252}Cf materials and technology. CIAE side built up a production line to produce ^{252}Cf neutron sources. It started to put into production in 1993. Up to now, there are more than 160 ^{252}Cf sources for industrial applications and not less than 20 ^{252}Cf sources for medical applications.

Neutron emission of ^{252}Cf source is only related to the amount of ^{252}Cf, so its fabricating technology can be simplified as the preparation of californium compound and the encapsulation of source capsules. Currently employed techniques, such as oxalate precipitation-calcination, porous material absorption, powder metallurgy-rolling and molecular plating are used in the preparation of neutron sources. The absorption of porous ceramic was adopted in China. The porosity of Al_2O_3 ceramic is about 70 %, and routine dimension is Φ 4.7 mm ×3.2 mm. In general, ^{252}Cf can be absorbed into ceramics from nitric acid feed solution (containing ~ 1 mg ^{252}Cf/mL), the active ceramic (source core) was rinsed with alcohol and dried at 100 ~ 105 ℃, then heated and calcined at 700 ℃ to convert the californium nitrate to a stable californium oxide (Cf_2O_3).

In order to ensure the safety, ^{252}Cf source core is doubly encapsulated in stainless steel capsule sealed by TIG welding. The regular ^{252}Cf neutron sources are obtained through decontaminating (using supersound washing), leakage testing and neutron monitoring. Neutron output of routine products are from 10^5 n/s to 10^9 n/s, The specification of ^{252}Cf neutron sources has Φ 7 mm × 15 mm or Φ 7 mm × 25 mm (containing 400 μg ^{252}Cf) for industrial uses, and has Φ 3 mm × 11 mm (containing 750 μg ^{252}Cf) for medical uses.

The characteristics of normal radioisotope neutron sources made in China are listed in Table 1. Fig. 7 ~ Fig. 10 give the neutron spectrum from ^{210}Po-Be source, ^{210}Po mock (^{235}U) fission source, ^{241}Am-Be source and ^{238}Pu-Be source respectively.

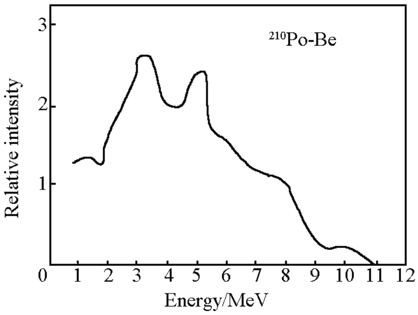


Fig. 7 ^{210}**Po-Be neutron spectra**

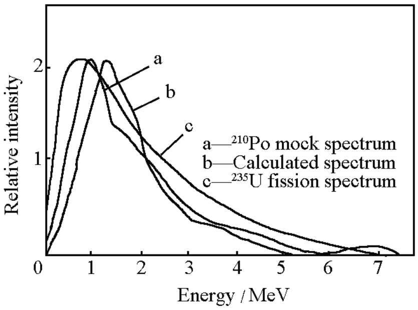


Fig. 8 ^{210}**Po mock fission neutron spectra**

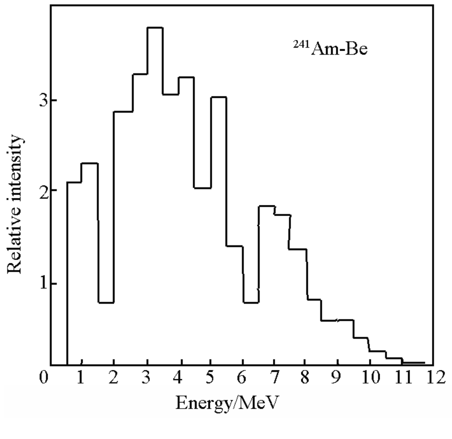


Fig. 9 ^{241} **Am-Be neutron spectra**

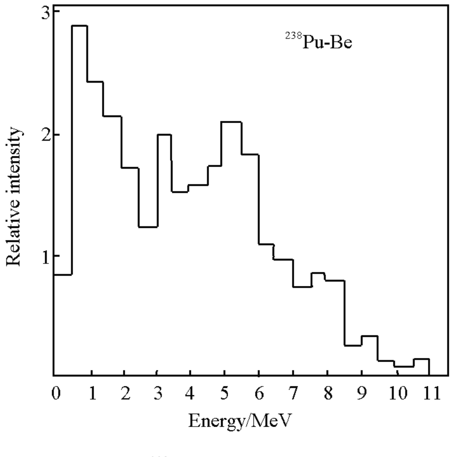


Fig. 10 ^{238}**Pu-Be neutron spectra**

Table 1 Characteristics of normal radioisotope neutron sources made in China

Neutron source	Half-life	Main energy /MeV	Neutron yield/(n• (s• Ci) $^{-1}$)	Neutron energy /MeV	
				E_{max}	E_{mean}
^{210}Po-Be	138.4 d	5.30 (α)	2.5×10^6	10.87	4.2
^{210}Po-Li	138.4 d	5.30 (α)	1.0×10^5	1.3	0.40
^{210}Po-LiF	138.4 d	5.30 (α)	3.1×10^5		0.585
Mock fission	138.4 d	5.30 (α)	$>1.2\times10^5$	10.87	1.7 ~ 1.9
^{241}Am-Be	433 a	5.48 (α)	2.2×10^6	11.5	4.0 ~ 5.0
^{238}Pu-Be	87.7 a	5.48 (α)	2.2×10^6	10.74	4.0 ~ 5.0
^{252}Cf	2.65 a	6.12 (α)	2.35×10^{12} (n/s• g)	12.8	2.3
^{124}Sb-Be	60.2 d	1.70 (γ)	1.5×10^5	0.024	

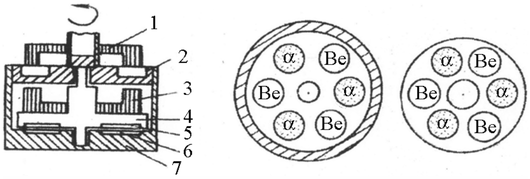
5 Special shape neutron sources

Most of normal radioisotope neutron sources were fabricated with cylindrical shape. In order to content the requirements of different applications, variable output neutron source (called switchable neutron source), annular neutron source, rod type neutron source (called source rod), spherical type neutron source and neutron needle were developed[39,40]. Several useful sources are described here.

5.1 Switchable neutron sources

In general, the neutron output is continuous for normal (α, n) neutron sources, because alpha emitter and target are mixed together. For certain applications such as neutron dosimeter, oil well logging and some physical tests, the development of new type neutron source is expected that neutron output is variable[41-43].

A special structure is adopted for the basic principle of switchable neutron source. The switch of neutrons output is performed through the closure or separation between α source and Be target[44]. ^{210}Po-Be switchable neutron source was developed by logging physical station of Daqing Oilfield and Department of Isotope, China Institute of Atomic Energy in 1975. The structure of source was shown in Fig. 11. Three pairs of ^{210}Po alpha sources (Φ 10 mm × 0.5 mm) and three pairs of Be discs (Φ 10 mm × 1.0 mm) were mounted on the bottom of capsule or the below of dial respectively. The total activity of ^{210}Po alpha sources is 114 GBq (3.08 Ci). Polonium is respectively electrodeposited on six gold foils. At beginning, ^{210}Po alpha sources and beryllium discs were arranged in such a way that it is possible to prevent the alpha emitter reaching target. Relying on the running magnets mounted on the inside and outside of sealed stainless steel capsule as well as JT-B type step relay, the change of opposite position of both ^{10}Po alpha source and Be disc were carried out.



1—Outer magnet; 2—Argon arc welding; 3—Inner magnet
4—Running spindle; 5—Be disc; 6—α source; 7—Capsule

Fig. 11 ^{210}Po-Be switchable neutron source

The test of prototype switchable neutron source indicated that the maximum neutron output was reached when the angle of running was 60°, 180° and 300°. The average neutron emission in the "on" position gave 7.30×10^5 n/s. The minimum neutron output was reached when the angle of running was 0°, 120°, 240° and 360° respectively. The average neutron emission in the "off" position gave 7.24×10^4 n/s. The neutron yields was 2.47×10^5 n/s• Ci. It indicated that neutron emission was approximately 10 % of conventional ^{210}Po-Be neutron sources. The ratio of neutron emission in "on" and "off" position was 10 ~ 11. The higher neutron emission in the "off" position was due to the rather close at edges of ^{210}Po alpha sources and Be targets.

5.2 Annular neutron sources

In order to meet the needs of neutron moisture gauge and neutron activation analysis, the new type of annular ^{241}Am-Be neutron sources have been developed since 1980. Comparing with the similar source abroad, there are some features in the aspects of structure design, source core preparing and sealing techniques.

It is very hard to fabricate a hollow cylindrical source, because the internal diameter of the source is limited by the dimension of BF_3 counter tube and the external diameter of the source must be kept to a reasonable size. These restrictions lead to a source design with a very small active volume. In addition, the distribution of active material should be homogeneous round the annulus and the neutron emission of the source should be stable.

An "inlaid structure" was adopted for the design of annular ^{241}Am-Be neutron sources (Fig. 12)[45]. The outer dimension of source is Φ 35 mm × Φ 26 mm × 18 mm and the dimension of annular void is Φ 33 mm × Φ28 mm × 9 mm. The pressed pellet technology was used in the preparation of source core (Amersham International Ltd. used "powder loaded" technology). The mixture of AmO_2 and beryllium powder was pressed into a hollow cylindrical source by double floating die (Fig. 13). Tests indicted that the density of active pellet was up to 1.2 g/cm^3 under the pressure of 4 t/ cm^2. The dimension of active pellet was Φ 33 mm × Φ 28 mm × 6 mm. It had enough mechanical strength and can content the demands of assembly. When the mass ratio of Be/AmO_2 is 12, the maximum loading amount of ^{241}Am can reach 0.8 Ci (~290 mg/Ci). So it is no problem to fabricate 50 ~ 500 mCi annular ^{241}Am-Be sources.

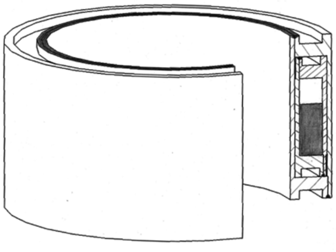


Fig. 12 Annular ^{241}Am-Be neutron source

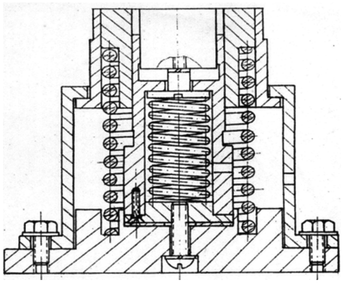


Fig. 13 Double floating die

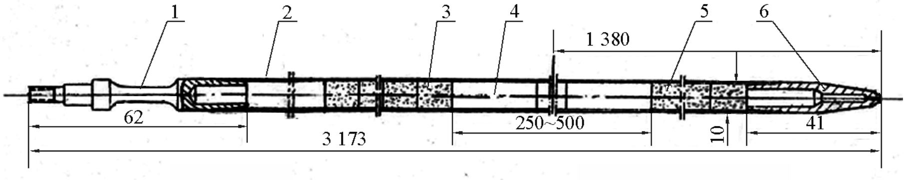
The annular ^{241}Am-Be neutron source is also doubly encapsulated in argon arc welded stainless steel capsule. Inner capsule (or inner annulus) and outer capsule (or outer annulus) are assembled respectively with inlaid structure . The numbers of weld for each capsule are decreased from 4 to 2 (Each end of capsule only has one weld). The total amount of weld is 4 (double annulus).

The advantages of new type annular ^{241}Am-Be neutron sources are of uniform americium distribution, stable neutron emission and excellent sealing quality. The innovation of the annular neutron source in structure is the subject of a China patent (ZL93207475.8)[46].

5.3 Neutron source rod

There are two kinds of start-up neutron sources for nuclear reactor. One is the "primary source" which used for initial cold clean start-up. ^{210}Po-Be source, ^{238}Pu-Be alloy source and ^{252}Cf source are often used as primary source, because they can meet the needs of small size and high neutron flux. Another is "second ary source" which is used for subsequent plant restart after extended shutdowns. ^{124}Sb-Be neutron sources are usually used as regenerative start-up sources for nuclear reactors.

In order to perform the start-up of pressurized water reactor (PWR) for the first nuclear power station in China, Qinshan Nuclear Power Corporation requested the supply of a pair of ^{210}Po-Be neutron source rods in 1989. The total neutron emission rates was more than 2×10^8 n/s, and the dimension of source rod was 10 mm in diameter and 3 173 mm long. The time of safety operation was about 450 d. Fig. 14 gives the structure and dimension of ^{210}Po-Be neutron source rod which has been completed. The initial drawing was designed by Shanghai Institute of Nuclear Engineering Research & Design. Passing through a large amount of tests, the drawing was modified for three times.



1—Top-end plug; 2—Clad tube; 3—Al_2O_3 spacer;
4—Neutron source; 5—Al_2O_3 spacer; 6—Bottom-end plug

Fig. 14 ^{210}Po-Be neutron source rod

The shape of start-up neutron source for nuclear power station is similar to fuel rod and control rod. It is named source rod. Slender in shape and high neutron emission are its features. Manufacturers encounter a challenge in techniques and equipments, because it is the first time to fabricate neutron source rods in China. In order to complete the task, the development of neutron source rod was conducted. The process of fabricating ^{210}Po-Be neutron source rods was composed of collection of polonium materials, preparation of source cores (^{210}Po-Be sources), encapsulation of source rods, packing and shipping.

For the core of source rod, its features are small physical size, high neutron emission and severe safety standard. In order to satisfy the requirements of a start-up neutron source rod, the design of ^{210}Po-Be source was considered both neutron yield and mechanical strength[47]. As a result of the calculation and comparison with different programs, the external dimension of capsule was 8.8 mm in diameter and 25 mm long (active area: Φ 5.6 mm × 16 mm). Each neutron source was sealed in double stainless steel (1Cr18Ni9Ti) capsules welded by TIG.

Depending on the stress calculation, the inner capsule can withstand helium pressure caused by the decay of polonium-210, (n, 2n), (n,α), (n,γ) nuclear reaction of beryllium as well as ambient temperature (~320 ℃) during the operation of PWR (453 d). Although the volume of active area was only 0.4 cm^3, the tests indicated that neutron yield can reach $(1.7 \sim 2.0) \times 10^6$ n/s. Neutron emission was $>1 \times 10^7$ n/s for each neutron source. Solid-solution 0Cr18Ni9Ti was selected as the material of capsules. It possesses higher mechanical strength, excellent radiation-resistant characteristics and better compatibility with cooling water (containing boron) of main loop. Prototype testing of 40 dummy sealed sources (no activity) indicated that the classification of safety performance was up to GB/E66545 (request: GB/E65545).

Sealing technology of ^{210}Po-Be neutron source rods are mainly involved in top-end plug girth welding, source core (^{210}Po-Be source) packing, bottom-end plug girth welding as well as end-closure (Fig. 15)[48]. For the key techniques, girth weld and end-closure weld were researched emphatically. Solid-solution 0Cr18Ni9Ti rod was selected as end-plug material and cold-worked 0Cr18Ni9Ti tube was selected as clad material (well thickness: 0.5 mm). The phenomena of micro-cracks within the melting zone for individual end-plug specimens were observed in the period of girth welding tests for jointing end-plug to the cladding. In order to eliminate this welded defects, a series of measures were taken, such as altering weld technology, welding parameters, cooling condition and inserted gape between the end-plug and the cladding. None of these methods had a major effect. However, obvious improvement was gained by using the end-plug with inner circumferential groove. Micro-cracks were eliminated owing to the melting metal partially entered into welding pool (inner groove). Finally, this project had been adopted. Helium arc welding was used in girth weld because of its good welding penetration and faster welding rate. The depth of fusion was controlled in 0.7 ~ 0.8 mm. All specimens passed a series of tests, such as tensile test, intergranular corrosion, metallographic test etc. The improvement of end-plug structure is the subject of a Chinese patent application (China patent: 92107230.9)[49].

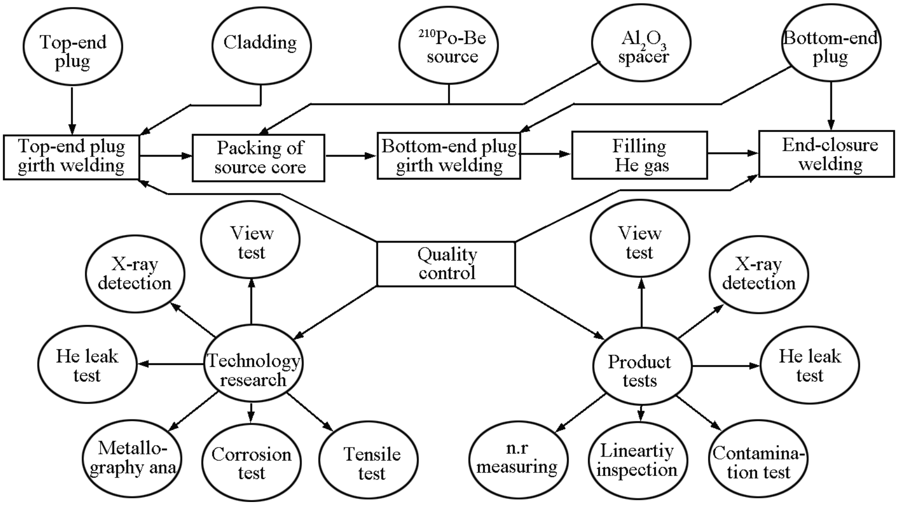


Fig. 15 Sealing process of ^{210}Po-Be neutron source rods

Another key technique is end closure of the bottom end-plug. It is imperative to have a cell to carry on the end closure welding, so as to keep the balance of pressure inside and outside of the rod. In addition, choosing the joint type of end-closure and the optimum welding parameters are also very important. In order to perform end closure of source rod, a small cell and a numerical-controlled spot welder were designed and manufactured. The helium arc spot welding was adopted to seal the end of bottom-end plug. Optimum type of end closure structure was selected through tests of 50 specimens. X-ray photography detection indicated that the depth of fusion was up to 2 mm and no defects were observed in the welding zone.

In order to perform the sealing of neutron source rods and prevent them from bending and deforming during the fabricating process, a versatile assembly line of neutron source rods was established (Fig. 16). It is about 12 m long collimated by laser. There are a lot of devices and facilities on the line. The fast neutrons from middle section of neutron source rod were shielded by movable twin water boxes. Major procedures can be accomplished on the same line. Finally, the source rod was transferred into paraffin container by remote control[50]. It is the biggest container (Φ 1 000 mm × 3 360 mm, 2.2 t weight) for packing radioisotope at home and satisfied the requests of neutron shield, safety transport and hoist a neutron source rod at Qinshan site.

Fig. 16 Assembly line of neutron source rods

Passing through near two years' work, a series of key techniques were solved. A pair of primary source rods (QS-1A rod and QS-1B rod) with ^{210}Po-Be neutron sources was made successfully on May 10, 1991. Total neutron emission was 3.0×10^8 n/s (± 5 %). Each rod contained 10 small ^{210}Po-Be neutron sources. The total activity for QS-1A rod and QS-1B rod was 3.78 TBq (102.2 Ci) and 3.80 TBq (102.7 Ci) respectively. All examinations demonstrated that the quality of source rods meets or exceeds the technical demands. A pair of ^{210}Po-Be neutron source rods had been put into the PWR core and performed the task of reactor start-up for the first nuclear power station in China.

Several pairs of ^{252}Cf neutron source rods were manufactured with present technology and facilities of source rod before and after new thousand years. They had been applied to Chashma Nuclear Power Station in Pakistan as well as other customers.

6 Prospect of radioisotope neutron sources in China

Owing to the urgent requirements of industry, agriculture, scientific research and national defense, the development of radioisotope neutron sources in China was promoted last century. And the developments of modern techniques including reactor start-up, oil well logging, moistrue measuring, activation analysis, process control, neutrond brachytherapy, neutron radiograph, etc were promote by the production of neutron sources and research of neutron characteristics, such as induced fission, neutron moderation, neutron capture, neutron activation, neutron attenuation, neutron scattering and neutron radiation[51]. Both social benefit and economic benefit are remarkable.

As a clean, safe and economic energy, nuclear power is a necessary selection. China nuclear electricity started in 1985. Eleven units had been set up by the end of 2004. Total installed capacity is up to 9.1 GWe. Depending on *The Long and Medium-term Nuclear Power Development Programme (2005 – 2020)*, modern China electricity generating reactors will deliver between 40 GWe and 50 GWe of electricity. In order to start-up reactors, it can be estimated that more then 30 pairs of primary neutron source rods and the same numbers of secondary neutron source rods will be needed within the next 15 years.

Radioactive well logger (FC-581) was firstly prepared by Xian Petroleum Exploration Instrument Complex (XPEIC). Later, more then one hundred scintillation radioactive well loggers (FC-651) were manufactured with domestic ^{210}Po-Be neutron sources (CIAE production) in the middle of 1960's[52]. They had been applied to Daqing, Shengli, Dagang oilfields, etc. Neutron well logging is used in the petroleum industry routinely for porosity determination and for differentiating oil and salt water. Owing to the porosity of rocks is served as reservoirs of oil and gas, the information of hydrogen contents can be obtained[53]. ^{210}Po-Be neutron sources were replaced by ^{241}Am-Be neutron source (~3 Ci) after using a decade. Several kinds of compensated neutron logging tool (CN-241, DG-1, BZC-1B, HN-CNL-8331) entered into market in the middle of 1990's[54]. These domestic instruments utilized high activity ^{241}Am-Be neutron sources or ^{238}Pu-Be neutron sources (18 ~ 20 Ci) provided by CIAE. Radioisotope neutron source played an important role in the exploration and exploitation of oil field in China over the past four decades. From now on, the amount of long-lived neutron sources will increase with the demands of nation economy for petroleum and mineral resources.

The first neutron moisture gauge (SHD-1) was fabricated by Department of Physics, Nanjing University in 1978[55]. This instrument used ^{241}Am-Be neutron source made in CIAE. Since then, the development of moisture measuring technique by neutron is very fast. Different kinds of neutron moisture gauges have been increased and the range of application has been extended. These instruments can be applied to analyze the moisture of coke, sinter, concrete, glass, pottery and fertilizer on line in industry. The quality of construction for highway, railway, airport and dam can also be examined by neutron moisture gauge. In the agriculture, the water measuring on surface layer and deep layer of soil can provide the scientific basis for saving water irrigation and soil improvement. In the coming days, the application scope of neutron moisture gauge will extend.

The first californium neutron afterloading system (ZH-1000 Neutron Knife) was manufactured by Shenzhen Linden Science & Technology Develop Co., Ltd. in 1998[56]. It has been applied to the clinic since 1999. Trials indicate that the intracavitary brachytherapys of ^{252}Cf fast neutron emitting are effective for uterine cervical cancer, endometrial cancer, esophageal cancer, rectal cancer, cardiac cancer etc. and the toxicity is lower. At present, there are 12 treatment centers with neutron knife equipments in China. About 30 cancer treating centers will utilize neutron knife in the near future. So the amount of medical ^{252}Cf neutron source will continue to increase.

In addition, neutron activation analysis as a high sensitivity, multi-elements and non-destroy analysis technique has been widely used in geology, metallurgy, industrial process control and environmental monitoring[57-59]. For instance, the automatic monitoring and match feed system of cement crude (using 10^7 n/s ^{241}Am-Be neutron source) have been used to measure the light elements: Al, Si, Mg, etc. Moreover, neutron radiography is a useful nondestructive test technique. The results from China University of Science & Technology indicate that ^{241}Am-Be neutron sources have the potentiality developing mini-detective system[60].

The supply of ^{241}Am or ^{238}Pu materials will be the bottle-neck of developing radioisotope neutron source in China. It is imperative to recover uranium and plutonium in the reprocessing of PWR spent fuel. At the same time, the recovery of ^{237}Np should be considered, so as to further produce ^{238}Pu. In addition, the extraction of ^{241}Am and other useful fission products from high-level liquid waste should be also considered[61].

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