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Development of ^{99}Mo Production Technology with Solution Irradiation Method

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1. Introduction

Technetium-99m (^{99m}Tc, half-life: 6.01 hours) is the world's most widely used radiopharmaceutical for exams of cancer, bowel disease, brain faculty and so on, and it is used for more than twenty million exams per year in the world and more than one million exams per year in Japan. The demand for ^{99m}Tc is continuously growing up year by year. The features of ^{99m}Tc as the radiopharmaceutical are as follows:

1. It is easy to add ^{99m}Tc to diagnostic medicines.
2. It is easy to measure the γ-ray energy with 0.14 MeV generated by isomeric transition from outside the body.
3. β rays are not emitted.
4. The patients' exposure associated with the exams is kept to the minimum because of the short half-life.

The production of the short-lived ^{99m}Tc is conducted by extracting from molybdenum-99 (^{99}Mo, half-life: 65.94 hours), which is the parent nuclide of ^{99m}Tc. Therefore, the stable production and supply of ^{99}Mo is very important in every country. All of ^{99}Mo used in Japan is imported from foreign countries. However, a problem has emerged that the supply of ^{99}Mo is unstable due to troubles in the import and the aging production facility (Atomic Energy of Canada Limited [AECL], 2007, 2008). In order to solve the problem, the establishment of an efficient and low-cost ^{99}Mo production method and the domestic production of ^{99}Mo are needed in Japan.

As a major ^{99}Mo production method, the fission method ((n, f) method) exists, and as a minor ^{99}Mo production method, the neutron capture method ((n, γ) method) exists. In order to apply to the Japan Materials Testing Reactor (JMTR) of the Japan Atomic Energy Agency (JAEA), two types of ^{99}Mo production methods based on the (n, γ) method have been developed in JAEA (Inaba et al., 2011): one is a solid irradiation method, and the other is a solution irradiation method, which was proposed as a new ^{99}Mo production technique (Ishitsuka & Tatenuma, 2008).

The solution irradiation method aims to realize the efficient and low-cost production and the stable production and supply of ^{99}Mo, and the fundamental research and development for the practical application of the method has been started (Inaba et al., 2009).

In this paper, a comparison between ^{99}Mo production methods, an overview of the solution irradiation method containing the structure of ^{99}Mo production system with the method and the progress of the development made thus far, estimates of ^{99}Mo production with the method, and the results of a newly conducted test are described.

2. Comparison between ^{99}Mo production methods

A comparison between the three ^{99}Mo production methods (the fission method, the solid irradiation method and the solution irradiation method) is shown in Table 1, assuming the ^{99}Mo production in JMTR, which is a tank-type reactor.

2.1 Fission method ((n, f) method)

In the conventional fission method ((n, f) method), high-enriched uranium targets are irradiated with neutrons in a testing reactor, and ^{99}Mo is produced by the ^{235}U (n, f) ^{99}Mo reaction. Most of the world supply of ^{99}Mo is produced by the (n, f) method since ^{99}Mo with a high-level specific activity of 370 TBq/g-Mo is obtained. However, the method has problems about the nuclear nonproliferation and the generation of a significant amount of radioactive waste including Fission Products (FPs) and Pu. Caused by the radioactive waste, the separation process of ^{99}Mo is too complex, and ^{99}Mo production with the (n, f) method needs expensive facilities and extreme care to avoid contamination with FPs. The ^{99}Mo production cost by this method achieves 57 US$/37 GBq (Boyd, 1997), and it is too expensive.

2.2 Neutron capture method ((n, γ) method)

2.2.1 Solid irradiation method

In the conventional solid irradiation method, solid targets including natural molybdenum such as MoO_3 pellets are irradiated with neutrons in a testing reactor, and ^{99}Mo is produced by the ^{98}Mo (n, γ) ^{99}Mo reaction. The post-irradiation process is only dissolution of the irradiated solid targets with an alkaline solution, and only a small amount of radioactive waste is generated in the process compared with the (n, f) method. The ^{99}Mo production cost of this method or the (n, γ) method is only 0.83 US$/37 GBq (Boyd, 1997).

However, the (n, γ) method has the disadvantage of producing ^{99}Mo with a low-level specific activity of 37-74 GBq/g-Mo and therefore the method has not had practical application in earnest. In order to utilize ^{99}Mo with the low-level specific activity, a high-performance adsorbent for (n, γ) ^{99}Mo is needed. The Japan Atomic Energy Research Institute (the present organization: JAEA) and KAKEN Inc. had developed the high-performance molybdenum adsorbent of Poly-Zirconium Compound (PZC) in 1995 (Hasegawa et al., 1996) and improved PZC (Hasegawa et al., 1999), and then the practical application of the (n, γ) method is just in sight. The molybdenum adsorbent performance of PZC is over 100 times compared with the conventional molybdenum adsorbent of alumina.

^{99}Mo production in JMTR will start by using the solid irradiation method. JMTR aims to provide ^{99}Mo of 37 TBq/w (1,000 Ci/w), and it will cover about 20% of the ^{99}Mo imported into Japan (Inaba et al., 2011).

2.2.2 Solution irradiation method

In the new solution irradiation method, a solution target including natural molybdenum such as an aqueous solution of a molybdenum compound (aqueous molybdenum solution) is irradiated with neutrons in a testing reactor, and ^{99}Mo is produced by the ^{98}Mo (n, γ) ^{99}Mo reaction. This new method is the improved type of the solid irradiation method, and it is possible to enhance the ^{99}Mo production compared with the solid irradiation method. The solution irradiation method has the following advantages compared with the solid irradiation method:

1. It is easy to increase the irradiated volume by using a capsule with larger volume than that of a rabbit. The rabbit is a small sized (150 mm length) capsule (Inaba et al., 2011).
2. The separation and dissolution processes after the irradiation are not necessary because the irradiation target is an aqueous solution.
3. The amount of generated radioactive waste is smaller than that of the solid irradiation method.

Items	^{99}Mo production methods		
	Fission method ((n, f) method)	Neutron capture method ((n, γ) method)	
		Solid irradiation method	Solution irradiation method
<Irradiation target>	Enriched ^{235}U	Natural Mo	Natural Mo
•Chemical type	•U-Al alloy, UO_2	•MoO_3, metal Mo	•Molybdate
•Form	•Foil, pellet	•Powder, pellet, metal	•Aqueous solution
•Quality control	•Complex	•Complex	•Simple
<Irradiation container>	Rabbit (30 cm^3)	Rabbit (30 cm^3)	Capsule (1,663 cm^3)
<Irradiation>			
•Pre-process of irradiation	•Adjustment of target and enclosing with container	•Adjustment of target and enclosing with container	•Adjustment of target
•Reaction of ^{99}Mo production	•^{235}U (n, f)	•^{98}Mo (n, γ) ^{99}Mo	•^{98}Mo (n, γ) ^{99}Mo
•Irradiation time	•5-7 days	•5-7 days	•5-7 days
•Collection of target	•Batch collection	•Batch collection	•Continuous or batch collection
•Post-process of irradiation	•Isolation in hot lab. (Complex)	•Dissolution in hot lab. (Relatively simple)	•No special treatment
Generated ^{99}Mo			
•Specific activity	•370 TBq/g-Mo	•37-74 GBq/g-Mo	•37-74 GBq/g-Mo
•Activation by-product	•Quite many (^{131}I, ^{103}Ru, ^{89}Sr, ^{90}Sr, etc.)	•^{92m}Nb	•^{92m}Nb, ^{14}C, ^{42}K, etc. (depending on a target solution)
<Mo adsorbent>	Alumina (2 mg-Mo/g-Al_2O_3)	PZC (250 mg-Mo/g-PZC)	PZC (250 mg-Mo/g-PZC)
<Radioactive waste>	Rabbits with FPs and Pu (Generation every one irradiation)	Rabbits (Generation every one irradiation)	Capsule (Lifetime: about 15 operation cycles)
<Production in Japan>	Difficult	Possible	Possible

Table 1. Comparison between three ^{99}Mo production methods

In this new method, efficient and low-cost ^{99}Mo production compared with the conventional ^{99}Mo production can be realized by using the (n, γ) reaction and PZC. This new method aims to provide 100% of the ^{99}Mo imported into Japan.

3. Overview of solution irradiation method

3.1 Structure of ^{99}Mo production system with solution irradiation method

The schematic diagram of the ^{99}Mo production system with the solution irradiation method is shown in Fig. 1. The system consists of an irradiation system, a supply and circulation system, and a collection and subdivision system. In the irradiation system, an aqueous molybdenum solution in a capsule installed in a reactor core is irradiated with neutrons under static or circulation condition, and ^{99}Mo is generated. In the supply and circulation system, the solution is supplied to the capsule through pipes and is circulated by a circulator in irradiation operation. A gas disposal device and a heat exchanger are installed in order to take measures against the radiolysis gas and heat generated from the solution by irradiation. The system is designed so as to minimize unirradiated solution. In the collection and subdivision system, after the solution including the generated ^{99}Mo is collected from the capsule through pipes, it is treated so as to be products such as PZC-^{99}Mo columns or ^{99}Mo transport containers.

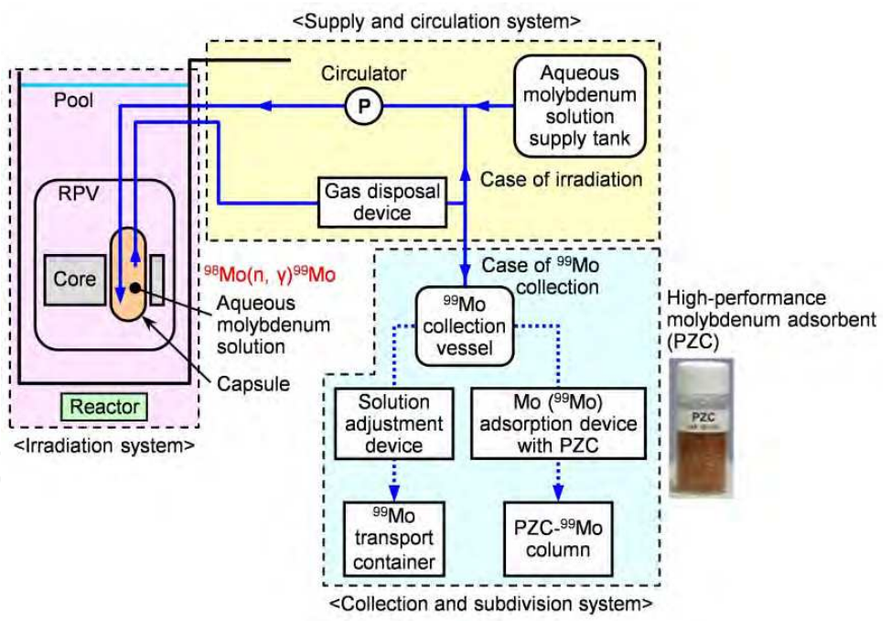

Fig. 1. Schematic diagram of ^{99}Mo production system with solution irradiation method

The detailed design of the ^{99}Mo production system is carried out based on the results of future investigations and tests.

3.2 Progress of the development made thus far

The most important element of the solution irradiation method is the aqueous molybdenum solution as the irradiation target. The solution with a high concentration near the saturation

is used for efficient ^{99}Mo production, and the solution always is in contact with the structural materials of the capsule and the pipes in the ^{99}Mo production system under irradiation. Aqueous molybdate solutions are promising candidates for the irradiation target. The effect of the solutions on metals such as the structural materials has been researched, and molybdates are known as corrosion inhibitors (Kurosawa & Fukushima, 1987; Lu et al, 1989; McCune et al, 1982; Saremi et al, 2006). However, the behavior of aqueous molybdenum solutions including the aqueous molybdate solutions under such the conditions is not well understood. Therefore, the following subjects about the fundamental characteristics of the solutions should be investigated:

1. Selection of the aqueous molybdenum solutions as candidates for the irradiation target
2. Compatibility between the solutions and the structural materials
3. Chemical stability of the solutions
4. Effect of γ ray and neutron irradiation on the solutions such as the radiolysis, the γ heating and the activation by-products.

The some subjects described above had already investigated (Inaba et al., 2009), and the progress of the development made thus far is explained as below:

3.2.1 Selection of candidates for irradiation target

The selection of candidates for the irradiation target was carried out. The conditions required for the irradiation target solution are as follows:

1. The irradiation target solution has the high molybdenum content for the efficient production of ^{99}Mo.
2. Few activation by-products are generated by target solution irradiation for the prevention of radioactive contamination.
3. The solution has good compatibility with the structural materials of the capsule and the pipes for the prevention of corrosion.
4. The solution is chemically stable and has no generation of precipitation for the prevention of an obstruction to the solution's flow.

Based on the conditions (1) and (2), two aqueous molybdate solutions (aqueous ammonium molybdate and potassium molybdate solutions) were selected as the candidates for the irradiation target among the aqueous solutions of general molybdenum compounds.

The solubilities of ammonium molybdate ($(NH_4)_6Mo_7O_{24}\cdot 4H_2O$) and potassium molybdate (K_2MoO_4) for pure water are 44 g/100 g-H_2O and 182.4 g/100 g-H_2O respectively, and the molybdenum contents in the solubilities of $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 are 23.9 g and 73.5 g respectively.

The activation by-product of $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ is only ^{92m}Nb. The activation by-products of K_2MoO_4 are ^{42}K and ^{92m}Nb. The γ-ray energy emitted from ^{42}K is high. However, by using PZC, it is possible to remove ^{42}K and ^{92m}Nb from the two aqueous molybdate solutions irradiated with neutrons.

The conditions (3) and (4) were confirmed by tests with the two solutions.

3.2.2 Unirradiation and γ-ray irradiation tests

Unirradiation and γ-ray irradiation tests were carried out by using the selected two aqueous molybdate solutions (aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 solutions), and compatibility between the two solutions and the structural materials of stainless steel and aluminum, the chemical stability, the circulation characteristics, the radiolysis and the γ heating of the two solutions were investigated. In addition, the integrity of PZC was investigated under γ-ray irradiation. As a result, the following were found:

1. The compatibility between the two static aqueous molybdate solutions and stainless steel is very well under unirradiation and γ-ray irradiation.
2. The two solutions are chemically stable and have smooth circulation under unirradiation and γ-ray irradiation.
3. The ratios of hydrogen in the gases generated by the radiolysis of the two solutions are higher than that of pure water.
4. The effect of γ heating on the two solutions is the same level as that on pure water.
5. The integrity of PZC is maintained under γ-ray irradiation.

However, the pH of the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution needs to be adjusted from weak alkaline to weak acid for the prevention of precipitation. This is a disadvantage as one of the candidates for the irradiation target.

At present, the aqueous K_2MoO_4 solution, which has no pH adjustment and has higher molybdenum content than that of the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution, is investigated as the first candidate of the irradiation target.

4. Estimates of ^{99}Mo production rates by solution irradiation method

The estimates of ^{99}Mo production rates by the solution irradiation method are shown in cases using aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 solutions as irradiation targets, assuming the ^{99}Mo production in JMTR.

4.1 Conditions for estimates of ^{99}Mo production rates

The JMTR core arrangement is shown in Fig. 2. The capsule of the ^{99}Mo production system with the solution irradiation method is installed into the irradiation hole, M-9 with maximum and average thermal neutron fluxes of 3.5×10^{18} n/(m$^2\cdot$s) and 2.6×10^{18} n/(m$^2\cdot$s) (Department of JMTR Project, 1994). The capsule consists of inner and outer tubes, and an aqueous molybdate solution is irradiated with neutrons in the inner tube to prevent the solution from leaking into the reactor coolant. Table 2 shows the conditions of the capsule and the two irradiation targets of the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 solutions.

^{99}Mo production rates are estimated based on the following conditions in addition to the conditions of Table 2:

- The generation and reduction of ^{99}Mo by the neutron capture reaction of ^{98}Mo (n, γ) ^{99}Mo and ^{99}Mo (n, γ) ^{100}Mo and the radioactive decay of ^{99}Mo are evaluated.
- ^{99}Mo doesn't exist in the initial stage of the calculation.
- The decay of neutron flux due to the inner and outer tubes of the capsule is considered.
- The circulation of the two irradiation targets is not considered.

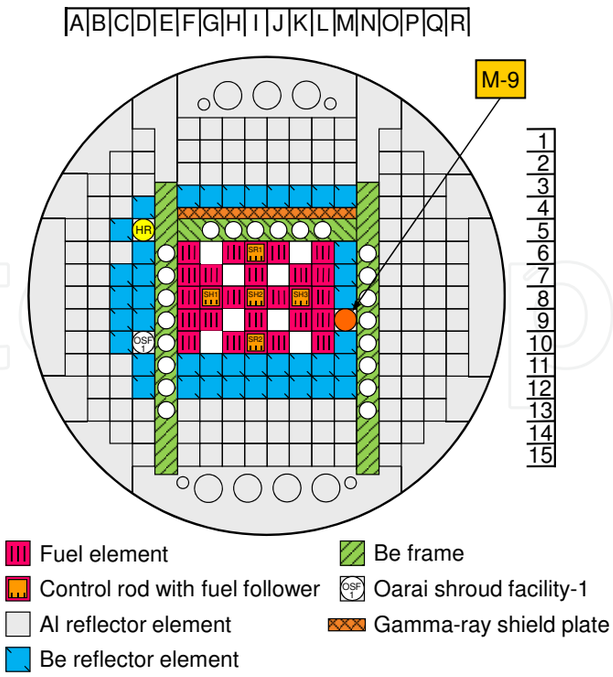

Fig. 2. JMTR core arrangement

Size and material of capsule	
Outer tube:	outer diameter of 65 mm, inner diameter of 61 mm
Inner tube:	outer diameter of 59 mm, inner diameter of 55 mm
Irradiation height:	700 mm
Irradiation volume:	1,663 cm^3
Material:	Stainless steel
Dissolved molybdenum in each irradiation target	
• Aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution (concentration: 90% of saturation): 372.8 g/1,663 cm^3 • Aqueous K_2MoO_4 solution (concentration: 90% of saturation): 702.7 g/1,663 cm^3	

Table 2. Conditions of capsule and two irradiation targets of aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 solutions

4.2 Basic equations for estimates of ^{99}Mo production rates

The disintegration rates of ^{98}Mo (isotopic ratio: 24.138%) and ^{99}Mo are shown as following equations:

$$\frac{dN_{98}}{dt} = -\phi\sigma_{98}N_{98} \tag{1}$$

$$\frac{dN_{99}}{dt} = -\left(\lambda + \phi\sigma_{99}\right) N_{99} + \phi\sigma_{98} N_{98} \tag{2}$$

The solutions of the equations (1) and (2) are as follows:

$$N_{98}(t) = N_{98}(0)\exp\left(-\phi\sigma_{98} t\right) \tag{3}$$

$$N_{99}(t) = \frac{\phi\sigma_{98}}{\lambda + \phi\left(\sigma_{99} - \sigma_{98}\right)} N_{98}(0)\left[\exp\left(-\phi\sigma_{98} t\right) - \exp\left\{-\left(\lambda + \phi\sigma_{99}\right)t\right\}\right] \tag{4}$$

where N_{98} and N_{99} are the atom number densities of ^{98}Mo and ^{99}Mo (n/cm^3), t is time (s), ϕ is neutron flux (n/(cm^2·s)), σ_{98} and σ_{99} are the capture cross section of ^{98}Mo and ^{99}Mo (cm^2), and λ is the decay constant of ^{99}Mo (1/s). When the neutron flux, the capture cross section, the decay constant and the time are given for the equations (3) and (4), ^{99}Mo generation rate per unit volume can be calculated depending on the time.

The specific activity of the generated ^{99}Mo is calculated from the following equation:

$$-\frac{dN_{99}}{dt} = \frac{W \times 4.17 \times 10^{23}}{AT} \tag{5}$$

where W is the mass of ^{99}Mo (g), A is the atomic mass number of ^{99}Mo, and T is the half-life of ^{99}Mo (s).

4.3 Estimated results of ^{99}Mo production rates

The relationship between the irradiation time and the calculated specific ^{99}Mo generation (generated ^{99}Mo activity per 1 g of molybdenum) is shown in Fig. 3. When the irradiation time is 6 days (144 h), the specific ^{99}Mo generation is 0.286 TBq/g-Mo as shown in Fig. 3.

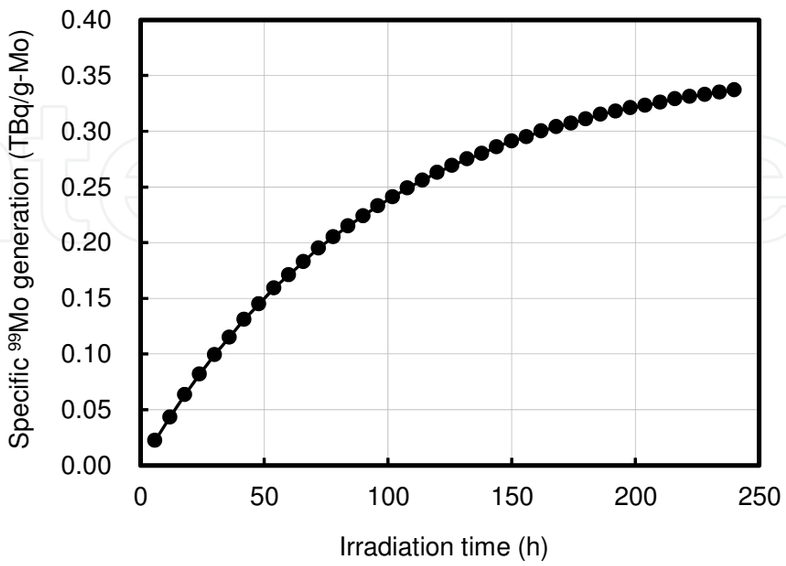

Fig. 3. Relationship between irradiation time and specific ^{99}Mo generation

Using the specific ^{99}Mo generation of 0.286 TBq/g-Mo, ^{99}Mo production rates are estimated. In the case using the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution as the irradiation target,

(^{99}Mo production in the case using the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution)

= 372.8 g × 0.286 TBq/g-Mo = 106.6 TBq = 2,881.9 Ci

In the case using the aqueous K_2MoO_4 solution as the irradiation target,

(^{99}Mo production in the case using the aqueous K_2MoO_4 solution)

= 702.7 g × 0.286 TBq/g-Mo = 201.0 TBq = 5,431.7 Ci

Here, the dilution effect by the unirradiated aqueous molybdate solution and the decay time of ^{99}Mo from the generation to the shipment are considered. It is assumed that the volume of the aqueous molybdate solution in the capsule and the pipes in the irradiation system and the supply and circulation system of the ^{99}Mo production system is about 2,500 cm^3 and that the time from the post-irradiation to the shipment is one day. After one day, ^{99}Mo decays to 0.78 times. Time from the irradiation to the shipment is one week. The ^{99}Mo production rates at the shipment are estimated as follows:

(^{99}Mo shipping activity in the case using the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution)

= 2,881.9 Ci × 1,663/2,500 × 0.78 = 1,495.3 Ci/w

(^{99}Mo shipping activity in the case using the aqueous K_2MoO_4 solution)

= 5,431.7 Ci × 1,663/2,500 × 0.78 = 2818.3 Ci/w

The ^{99}Mo production rate in the case using the aqueous K_2MoO_4 solution is about twice compared with that in the case using the aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ solution. It is a distinct advantage of the aqueous K_2MoO_4 solution. However, in order to aim to provide 100% of the ^{99}Mo (5,000 Ci/w) imported into Japan and to increase the production rate, some ideas such as the concentration of ^{98}Mo are needed.

5. Compatibility test between flowing aqueous molybdate solution and structural material

In the ^{99}Mo production system with the solution irradiation method, a flowing target solution with a high concentration is in contact with the structural material of the capsule and the pipes, and then it is important to investigate compatibility between the flowing target solution and the structural material. In the previous tests (Inaba et al., 2009), the circulating solution test was carried out under γ-ray irradiation. However, the SUS304 specimen used in the test was only immersed in the bottom of an irradiation container with a volume of 2,000 cm^3, and the specimen had no influence of the circulating solution flow, and then the compatibility was not cleared. Therefore, the compatibility test between the flowing target solution and the structural material was carried out, and the corrosivity of the flowing target solution for the structural material as well as the chemical stability of the solution was investigated.

An aqueous K_2MoO_4 solution, which was the first candidate of the irradiation target, was used in the test. The purity of K_2MoO_4 used in the test was over 98%.

5.1 Test apparatus

Fig. 4 shows the schematic diagram of a test apparatus, which was used in order to investigate compatibility between a flowing aqueous K_2MoO_4 solution and a structural material and the chemical stability of the solution. The test apparatus consists of a immersion container for immersing specimens under flow, a glass storage tank with a volume of about 700 cm^3, a thermocouple inside the storage tank for solution temperature measurement, a feed pump to circulate the solution, a flowmeter, Teflon tubes with an inner diameter of 7.5 mm to connect each component, two syringes, which were used for depressurization, solution supply, air purge and solution sampling, a data logger to collect temperature data and to monitor the temperature and so on. Some components such as the immersion container and the storage tank were installed into a heating chamber to heat the solution.

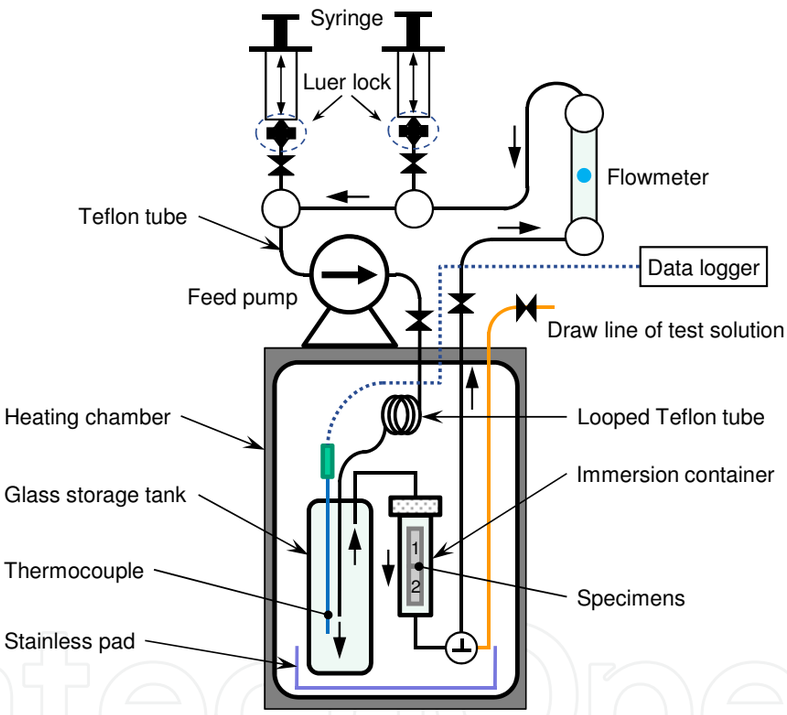

Fig. 4. Schematic diagram of test apparatus for compatibility test

The immersion container consists of a glass outer tube with an outer diameter of 22 mm and a height of 62.5 mm and a Teflon inner holder with an inner diameter of 13 mm and a height of 60 mm, and two specimens (specimen 1 and 2) were fixed in the center of the container by the holder as shown in Fig. 5 and they were arranged one above the other in the container. The storage tank was located upstream of the immersion container to keep the solution temperature constant and to prevent the solution from pulsating by the feed pump. In addition to the storage tank, a looped long Teflon tube connected between the pump and the storage tank was used to keep the solution temperature in the heating chamber. The total length of the circulation route of the solution was about 6.8 m, and the total quantity of circulating solution was about 300 cm^3 except the volume of the storage tank.

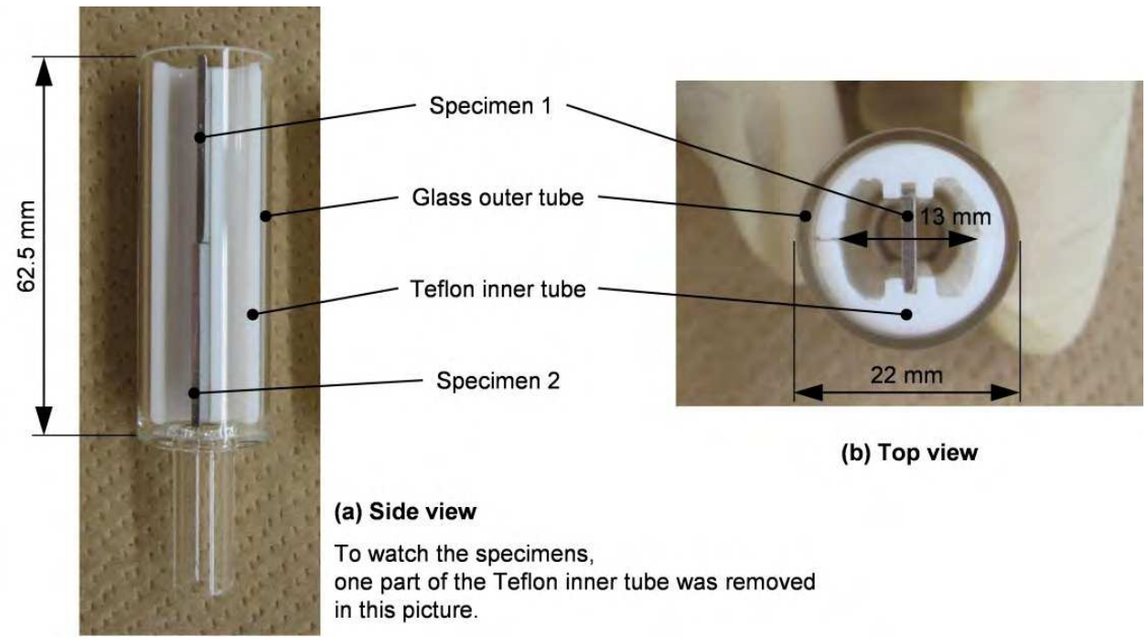

Fig. 5. Structure of immersion container

5.2 Test method and conditions

The compatibility test was carried out by using the test apparatus with a closed loop shown in Fig. 4. After the specimens were set in the immersion container, aqueous K_2MoO_4 solution was injected in the closed loop, and the solution was circulated at a constant flow rate. The flow rate was set at about 120 cm^3/min, considering the flow velocity assumed in an actual ^{99}Mo production system. The concentration of the solution was adjusted to about 90% of the saturation for the prevention of crystallization, and the temperature of the solution was maintained at about 80˚C for the prevention of boiling. As the specimens immersed in the solution, stainless steel SUS304 was used based on the results of the previous immersion tests (Inaba et al., 2009). SUS304 has been used as the structural material of capsules and pipes in JMTR. The size of the specimens was $10^{W}\times30^{L}\times1.5^{T}$ mm. Table 3 shows the chemical composition of a SUS304 specimen. The total immersion time of the specimens was 112.7 days, and the immersion time under flow was 84.5 days out of a total of 112.7 days. The total immersion time was longer than the immersion time under flow because the feed pump was temporarily stopped by the planned blackouts and the pump troubles.

During the test, at regular intervals, the specimen 1 was taken from the immersion container, and the specimen's weight was measured after washing by pure water and drying, and its surface state was observed. In addition, the aqueous solution was sampled from the closed loop by using one of the syringes, and the pH and molybdenum concentration of the solution were measured, and the solution state was observed.

After the test, the specimen 1 and 2 were taken from the immersion container, and the specimens' weight was measured, and their surface states were observed. In addition, the aqueous solution was sampled from the closed loop, and the pH and molybdenum concentration of the solution were measured, and the solution state was observed.

C	Si	Mn	P	S	Ni	Cr	Fe
0.06	0.51	0.73	0.026	0.002	8.03	18.07	Balance

(Unit: wt%)

Table 3. Chemical composition of SUS304 specimen

5.3 Results and discussions

The average temperature and flow rate of the aqueous K_2MoO_4 solution used in the test were 81˚C for a total immersion time of 112.7 days and 123 cm^3/min for a total immersion time under flow of 85.5 days respectively.

5.3.1 Corrosivity of flowing aqueous K_2MoO_4 solution for SUS304

The surface states of the two SUS304 specimens before and after the immersion in the flowing aqueous K_2MoO_4 solution for a total of 84.5 days are shown in the Fig. 6, and the relationships between the immersion time and corrosion rates of the specimens are shown in Fig. 7. The corrosion rates were estimated by the following equation:

$$Corrosion\ \ rate = \frac{Weight\ \ change}{Surface\ \ area \times Immersion\ \ time \times Density} \tag{6}$$

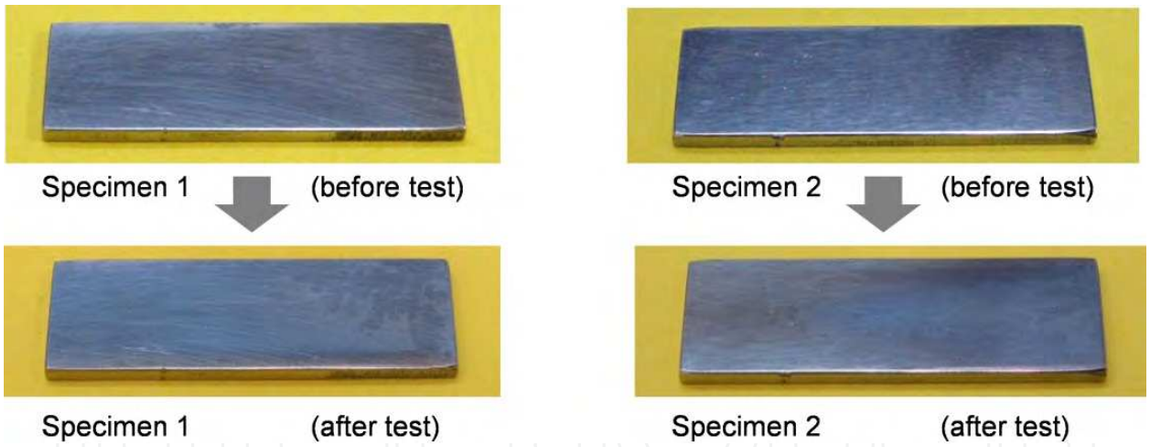

Fig. 6. Surface states of SUS304 specimens before and after compatibility test

The equation (6) shows the wastage thickness per unit time. In the visual observation and comparison of the two specimens' surfaces before and after the compatibility test, whereas streamlined patterns, partly slight tarnish and the partly slight loss of metallic luster were found on the surfaces, obvious corrosion such as corrosion products was not found. The corrosion rate of the specimen 1 increased temporarily to 0.10 mm/y in the initial stage of the test (an immersion time of 21 days) and decreased finally to 0.02 mm/y. On the other hand, the corrosion rate of the specimen 2 was 0 mm/y at the beginning and end of the test. There was no change in the state of the specimen 1 surface in the initial stage of the test, and the temporary increase of the specimen 1 corrosion rate might be affected by taking out from the immersion container.

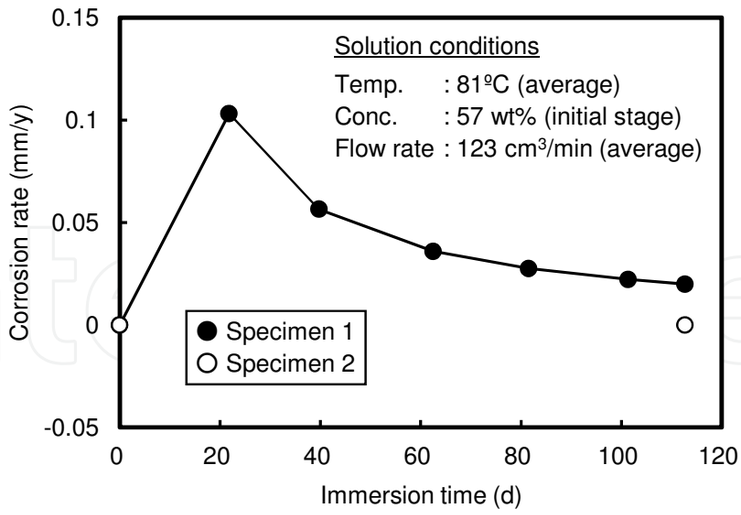

Fig. 7. Relationships between immersion time and corrosion rates of SUS304 specimens immersed in flowing aqueous K_2MoO_4 solution for 84.5 days

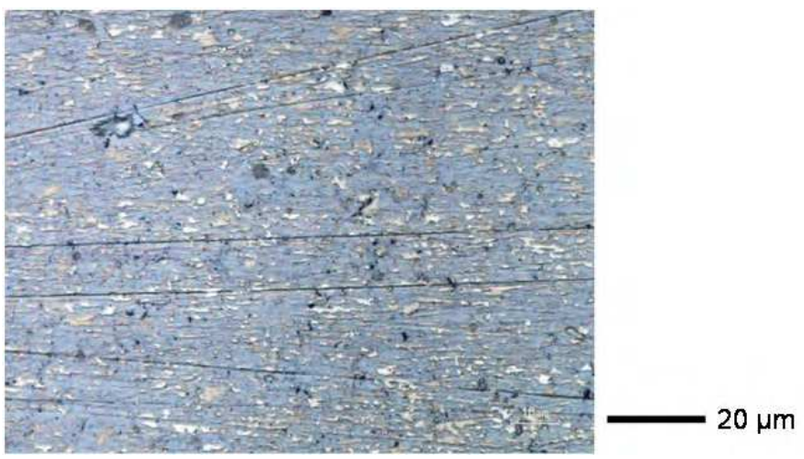

Fig. 8. Inverted materials microscope photograph of specimen 2 surface immersed in flowing aqueous K_2MoO_4 solution for 84.5 days

For the confirmation of the detailed surface states, the specimen 2 as the representative of the two specimens were observed and analyzed with an inverted materials microscope and a field emission Electron Probe Micro Analyzer (EPMA). Fig. 8 shows the inverted materials microscope photograph of the specimen 2 surface. The black lines and dots in Fig. 8 are preexistent scratches and hollows. Tarnish is recognized on the surface. Fig. 9 shows the Scanning Electron Microscope (SEM) photograph of the specimen 2 cross-section surface taken with the EPMA, and Fig. 10 shows the color map of the specimen 2 cross-section surface analyzed with the EPMA. The cross-section surface was prepared by cutting the center of the specimen 2, mounting in a resin and polishing. A thin coating layer, which is thought to be the cause of the tarnish, is found on the surface as shown in Fig. 9. To see Fig. 10, K and Mo, which are the main components of K_2MoO_4, are not detected and a relatively-high level of Si is detected on the surface. After the test, the corrosion of the glass outer tube in the immersion container was found, and then it is considered that the main component of

the coating layer is Si eluted from the tube. This Si coating layer might inhibit the corrosion of the specimens. In any case, the progress of the corrosion was not observed in the SUS304 specimens, and SUS304 has good compatibility with a flowing aqueous K_2MoO_4 solution.

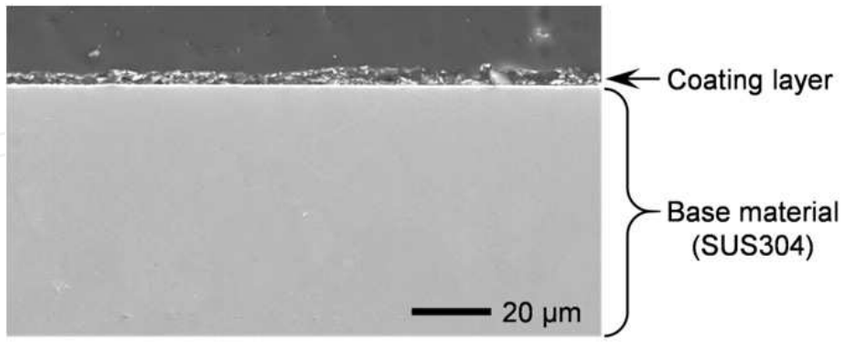

Fig. 9. SEM photograph of specimen 2 cross-section surface immersed in flowing aqueous K_2MoO_4 solution for 84.5 days

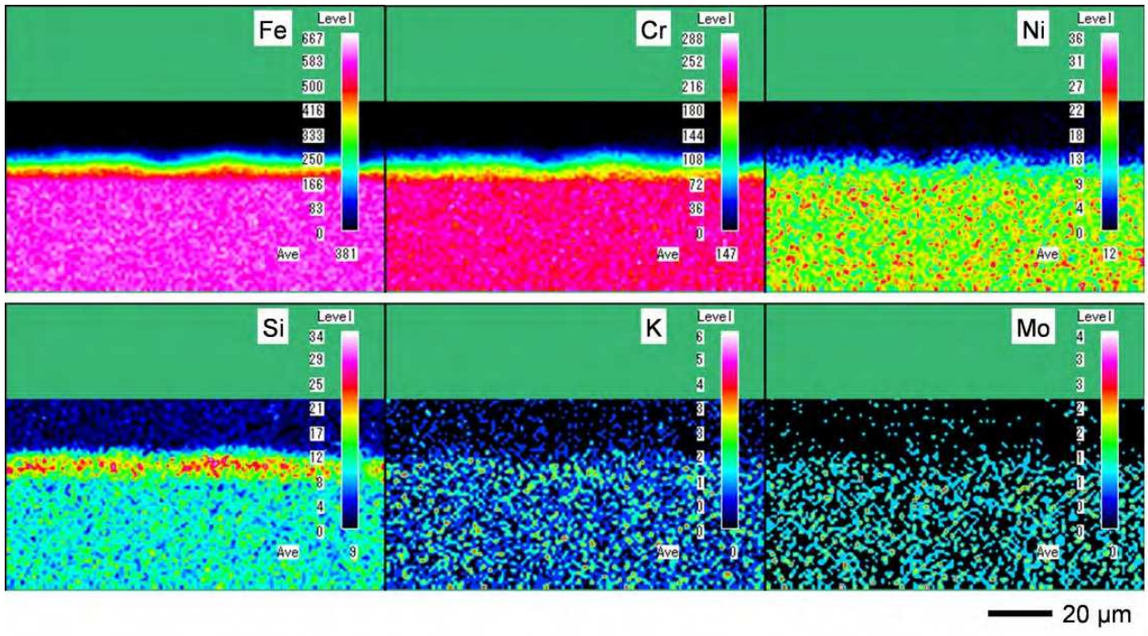

Fig. 10. EPMA color map of specimen 2 cross-section surface immersed in flowing aqueous K_2MoO_4 solution for 84.5 days

5.3.2 Chemical stability of flowing aqueous K_2MoO_4 solution

During the test term, the aqueous K_2MoO_4 solution was chemically stable, and the precipitation or the deposit was not generated in the solution. Then the molybdenum concentration of the solution was almost constant before and after the test, and the concentrations before and after the test were 396.2 mg/mℓ and 384.0 mg/mℓ respectively. The concentrations were measured with an Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES). The pH of the solution was also almost constant at pH9.5-9.7.

6. Conclusion

In the ^{99}Mo production system with the solution irradiation method, a static or flowing aqueous molybdenum solution in a capsule is irradiated with neutrons in a testing reactor, and ^{99}Mo is produced by the ^{98}Mo (n, γ) ^{99}Mo reaction. The system aims to provide 100% of the ^{99}Mo imported into Japan. As a part of the technology development, aqueous $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ and K_2MoO_4 solutions were selected as candidates for the irradiation target of the system, and compatibility between the static two solutions and the structural materials of the capsule and pipes in the system, the chemical stability, the radiolysis and the γ heating of the solutions were investigated. As a result, it was found that the solutions are promising as the target. In addition, compatibility between a flowing aqueous K_2MoO_4 solution, which was the first candidate for the irradiation target in terms of a ^{99}Mo production rate, and the structural material and the chemical stability of the flowing solution were investigated. As a result, it was found that stainless steel SUS304 has good compatibility with a flowing aqueous K_2MoO_4 solution and that the solution is chemically stable. The fundamental characteristics of the selected aqueous molybdate solutions became clear, and SUS304 can be used as the structural material of the capsule and the pipes.

In the future, a neutron irradiation test will be carried out as an overall test of ^{99}Mo production system with the solution irradiation method, and ^{99}Mo production, the separation of activation by-products, the quantity of radiolysis gas, nuclear heating and so on will be investigated.

Aiming at the domestic production of ^{99}Mo in Japan, the development of ^{99}Mo production with the solution irradiation method is kept going.

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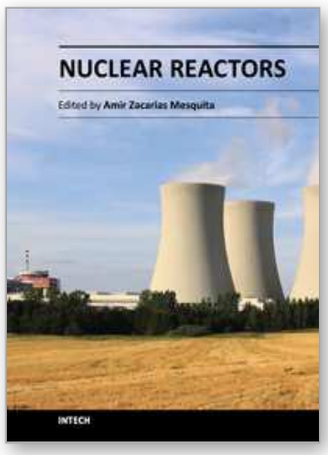

Nuclear Reactors

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This book presents a comprehensive review of studies in nuclear reactors technology from authors across the globe. Topics discussed in this compilation include: thermal hydraulic investigation of TRIGA type research reactor, materials testing reactor and high temperature gas-cooled reactor; the use of radiogenic lead recovered from ores as a coolant for fast reactors; decay heat in reactors and spent-fuel pools; present status of two-phase flow studies in reactor components; thermal aspects of conventional and alternative fuels in supercritical water?cooled reactor; two-phase flow coolant behavior in boiling water reactors under earthquake condition; simulation of nuclear reactors core; fuel life control in light-water reactors; methods for monitoring and controlling power in nuclear reactors; structural materials modeling for the next generation of nuclear reactors; application of the results of finite group theory in reactor physics; and the usability of vermiculite as a shield for nuclear reactor.

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