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After the feasibility study on ^{99}Mo production by (n,γ) method at HANARO was published by a KAERI report, worldwide supply of ^{99}Mo became worse and a need for early available alternative ^{99}Mo became stronger. Previous study indicated that the (n,γ)^{99}Mo has a potential to be an alternative mass ^{99}Mo available earlier than those by any other methods. It can be realized when radioisotope industry of each country accepts the use of (n,γ)^{99}Mo for a meaningful portion of national demand. A good backup supply system among high flux reactors in the region is a prerequisite to guarantee a stable and sufficient availability of the (n,γ)^{99}Mo for the region, for which active collaboration among reactors is essential. As the initial stage of collaboration between HANARO and JMTR for the (n,γ)^{99}Mo supply, the specific experience and ^{99}Mo production capability in HANARO have been discussed and revisited on the base of the previous report.

Keywords:Medical Diagnosis, ^{99}Mo Production, ^{99m}Tc, Backup Supply, HANARO, Neutron, Cross Section

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HANAROでの(n, γ)法による^{99}Mo製造に関する再評価

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HANAROでの(n, γ)法による^{99}Mo製造に関する可能性の検討が韓国原子力研究所(KAERI)により出版された後、世界的に^{99}Mo供給が不足し、早急に代替の^{99}Mo供給の必要性が要求されている。以前の研究から、(n, γ)法による^{99}Mo供給が他の製造法よりも早く大量製造に結び付く可能性があることが指摘されていた。各国の放射性同位元素産業が世界需要の重要な項目として(n, γ)法による^{99}Mo使用を容認された時、より現実なものとなる。各地域の高フラックス原子炉による供給システムは、(n, γ)法による^{99}Moの安定かつ十分な有効性を保証するための必要条件であり、原子炉間の協力が重要である。

本報告書は、(n, γ)法による^{99}Mo供給のためのHANAROとJMTRにおける最初の協力として、HANAROにおける経験と^{99}Mo製造容量を確認し、以前の報告書に基づいて再評価したものである。

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1. Introduction

A new adsorbent ALSUL [1] having a potential of ^{99m}Tc generators with (n,γ)^{99}Mo, the intention of JAEA [2,3] to produce (n,γ)^{99}Mo at JMTR, and construction of CARR (China Advanced Research Reactor), together with a shortage in ^{99}Mo supply over the world had motivated a feasibility study on the (n,γ)^{99}Mo production at HANARO. A KAERI report [4] was published as a result of the study.

It indicated that if an effective regional backup supply scheme with high flux reactors in Japan and China is established, HANARO can produce sufficient (n,γ)^{99}Mo for the region. Specific activity of (n,γ)^{99}Mo from HANARO, in combination with the ALSUL, was marginally enough to make the highest activity ^{99m}Tc generators under use in Korea. Therefore, it was expected that if a new ^{99m}Tc generator using ALSUL is fully verified, a large portion of ^{99m}Tc generators loaded with imported fission Mo could be replaced by the new generators loaded with (n,γ)^{99}Mo. There are a few high flux reactors in Japan and China. Therefore, if they also produce (n,γ)^{99}Mo, the regional backup supply scheme could be made for a stable availability of ^{99m}Tc in the region.

Since Japan imports all ^{99}Mo from long distance overseas countries, the impact from the instability of world ^{99}Mo supply is also large. JAERI [5] studied fission Mo production using enriched uranium from 1972 to 1977 and then (n,γ)^{99}Mo production until 1985. Studies on the (n,γ)^{99}Mo production [6] resumed rather recently for the ^{99}Mo supply when JMTR restarts after significant refurbishment. But commercial production by local reactors has never been experienced yet.

About 80% of ^{99}Mo in Japan is used by 500 Ci Master milkers with remaining 20% by portable generators, which is quite different from Korea. Therefore, technical challenge to replace fission Mo to (n,γ)^{99}Mo in Japan is also very different from Korea. Major target of JAEA is replacing the Master milkers by (n,γ)^{99}Mo rather than the portable generators. Anyway, both KAERI and JAEA are studying how to make the (n,γ)^{99}Mo effective for the stable availability of ^{99m}Tc in respective countries. If JMTR produces (n,γ)^{99}Mo, it will need backup reactors as well. Therefore, they are strong potential reactors for a cross backup supply of (n,γ)^{99}Mo in short distance.

The backup will be possible when the backup reactors also produce (n,γ)^{99}Mo for their own supply. Therefore, collaboration among potential backup reactors is very important for the realization of (n,γ)^{99}Mo production, and its improvement is needed. As the world ^{99}Mo supply has become worse after the publication of previous report, need for an alternative supply available within short time became strong desire. From such viewpoints, the feasibility of (n,γ)^{99}Mo production at HANARO is revisited. New reports reflect changes and progress afterward, and additional information worthwhile for the collaboration. General feasibility on mass (n,γ)^{99}Mo production will be published by another JAEA report [7]. This report narrows down to the specific experience of KAERI and $(n,\gamma)^{99}Mo$ production capability of HANARO.

As explained in Section 2, KAERI has a long experience in supply of ^{99m}Tc and studied various ways for its stable availability in Korea. However, its current availability is not so robust under the shortage of world ^{99}Mo supply. Together with other motive of study explained in Section 3, authors expect that the $(n,\gamma)^{99}Mo$ production at high flux reactors has strong potential to enhance the availability of ^{99m}Tc not only in Korea but also for other countries. In order to have an insight into its feasibility, $(n,\gamma)^{99}Mo$ production capability of HANARO was calculated for three selected irradiation holes and various targets as explained in Section 4. Section 5 analyzes its results to find out potential guidelines for effective ways of target irradiation, Section 6 evaluates production capability of HANARO, and Section 7 suggests an irradiation device for the production at HANARO.

There are uncertainties in the calculation and further refinement is needed for an actual production. However, it is clear that an irradiation hole located just outside of the core in HANARO can produce enough ^{99}Mo not only for Korea but also for the region which is defined in the Appendix.

2. Experience of KAERI relevant to ^{99}Mo

From long time back when two low power TRIGA reactors were only available in Korea, KAERI has contributed to domestic availability of several radioisotopes and relevant technical services, as well as encouraging their commercialization by local industry. Whenever any of them became available from commercial side with sufficient reliability for stable supply with reasonable price, KAERI had to discontinue its supply. The supply of ^{99m}Tc solution is an example.

KAERI had supplied ^{99m}Tc solution produced by $(n,\gamma)^{99}Mo$ in TRIGA reactors. In 1984, a convenient method for the extraction of ^{99m}Tc [8] was devised. When HANARO began power operation from end of 1995, two TRIGA reactors were permanently shut down and the $(n,\gamma)^{99}Mo$ production was transferred to HANARO. As domestic demand on ^{99m}Tc gradually increased, import of ^{99m}Tc generators increased and the portion of HANARO supply became small. However, the supply from HANARO was important for stability in ^{99m}Tc availability as well as its price.

When the Korean currency was suddenly depreciated to less than a half due to national economic difficulty from the end of 1997, the price of ^{99m}Tc generators jumped to above twice. Then many other hospitals who had been reluctant for the use of ^{99m}Tc solutions from HANARO requested the ^{99m}Tc supply. As the demand to HANARO expanded, 98.5% enriched $^{98}MoO_3$ was purchased from Russia in 1998 to increase the production effectively. For a continued stable supply in an expanded amount, even though its portion in domestic supply was still small, HANARO had to be operated by a weekly mode due to unavailability of backup supply from any other reactors. It was ended in 2003 when KAERI completed a ^{99m}Tc generator loading facility and, using the facility, a company commenced supply of ^{99m}Tc generators by importing fission Mo. After then, it seemed that HANARO would be free from the ^{99m}Tc supply.

Recently, however, severed shortage of ^{99}Mo worldwide requested HANARO to supply the ^{99m}Tc again. HFR announced 6 months shutdown from 19 February 2010 to repair leaking primary cooling pipes embedded in the reactor pool concrete, but the shutdown NRU to repair reactor vessel from heavy water leak was not ready to operate yet. As urgent responses announced by major ^{99}Mo suppliers, MARIA in Poland would start irradiation of fission Mo targets to backup the HFR, and operation schedules of BR2 and OSIRIS would be adjusted to minimize the impact from the long term shutdown of HFR. But a possibility of shortage in the ^{99}Mo supply in Korea had to be considered. Therefore, Korean Government called a meeting on 16 February 2010 to check the status of domestic ^{99}Mo supply [9]. The meeting concluded "*A weekly ^{99}Mo demand in Korea is 110 Ci, which has been satisfied by importing 90 Ci from SAFARI and 20 Ci from HFR. The 20 Ci from HFR will be replaced by the 90 Ci from SAFARI as much as possible, an effort will be made to import additional 20 Ci from BR2 and OPAL, and HANARO will supply 10 Ci with potential increase to 40 Ci by using enriched ^{98}Mo. In case SAFARI shutdowns, HANARO will be operated for the ^{99m}Tc supply with the highest priority.*" HANARO already supplied ^{99m}Tc when SAFARI shut downed during 17 ~ 23 November 2009. The meeting also recommended a study on a new research reactor in Korea for the stable availability of the radioisotopes.

Actually, keeping the device and skilled manpower for several years without any ^{99m}Tc supply is not practical. Fortunately, however, resuming the ^{99m}Tc supply from HANARO was possible because some activity on the device has been continued. The device had become renowned for an effective extraction of ^{99m}Tc from very low specific activity ^{99}Mo, and has been supplied to several countries especially through Technical Cooperation Programs of IAEA. Recently, a Russian company showed interested in this device and they visited KAERI for possible purchasing. Therefore, the device and manpower for its operation could be maintained, and thereby the ^{99m}Tc could be supplied when it was fully justified.

As already experienced before, however, if a large portion of ^{99m}Tc supply is needed for a long term, a significant burden will be loaded to the HANARO. The current situation of worldwide ^{99}Mo supply indicates possibility that the weekly operation of HANARO may recur. Much expanded use of the HANARO nowadays with many new expensive experimental facilities will make the weekly operation much more difficult than before.

While backup supply of ^{99m}Tc from nearby countries is neither possible due to its short life nor practical due to its daily supply feature, backup supply of (n,γ)^{99}Mo is possible by irradiated targets. Should the backup supply be available, above burden of HANARO can be unloaded. A reliable and stable backup supply is possible when multiple high flux reactors in nearby countries produce the (n,γ)^{99}Mo in a concerted mode under a well organized cross backup scheme.

However, in general, the ^{99m}Tc solution supply is not as convenient as the supply of ^{99m}Tc generators. While a generator is delivered to a hospital once a week, the ^{99m}Tc solution must be delivered very early in the morning every working day. It was also said that the ^{99m}Tc solution supplied from KAERI had given poor image for brain scanning. But there is neither document nor person understanding its reason. The team testing ALSUL intends to test this matter also. Anyhow, all ^{99m}Tc except from HANARO has been obtained from portable ^{99m}Tc generators, and it is certain that the radioisotope industry in Korea has preferred the supply of ^{99m}Tc generators rather than its solution. But generators using (n,γ)^{99}Mo have not been competitive with those using fission Mo.

KAERI has studied fission Mo production for long time. In 1995 when HANARO just started operation, USA Government indicated a possible supply of HEU (highly enriched uranium) for the ^{99}Mo production at HANARO. The USA already supplied HEU to Indonesia together with Cintichem technology. Therefore, KAERI was encouraged to initiate a project to produce fission Mo at HANARO. At that time, the MAPLE project which commenced early 1980s in Canada had been halted from 1992 though majority of reactor structures were completed. After then, the project resumed in 1996 and commissioning of the first MAPLE started in 2000. After then, when the HEU was actually needed for tests at HANARO, the USA refused its supply, which terminated the KAERI project.

KAERI has been participating in a Coordinated Research Project (CRP) of IAEA on ^{99}Mo production using LEU (low enriched uranium) or neutron activation [10]. Under the CRP, KAERI developed a technology producing LEU foils for targets and many uranium foils have been distributed to CRP member states for their tests. Meanwhile, feasibility on MIP (Medical Isotope Producer) using solution LEU fuel was studied, but information on construction of a MIP in Chengdu, China, discontinued it.

It was concluded that construction of any new facility for the fission Mo production at the current KAERI site is impractical. There have been debates on the construction of a new research reactor in Korea, of which function will certainly include the ^{99}Mo production as one of highest priorities. Very recently, Korean Government is considering a feasibility study on the construction of a new research reactor. If the reactor is built, it will contribute to the stable availability of ^{99}Mo either by the fission Mo or by the (n,γ)^{99}Mo, but at least six years will be needed until the ^{99}Mo is available from the reactor.

KAERI had participated in a FNCA program for a new generator technology development using (n,γ)^{99}Mo and PZC [11]. However, the study in KAERI discontinued after termination of the program because generators using (n,γ)^{99}Mo seemed no more needed because a company commenced operation of the ^{99m}Tc generator loading facility by importing fission Mo.

Rather recently KAERI developed a new high adsorbent [1] and named it ALSUL. The

ALSUL was initially developed to enhance the $^{188}W/^{188}Re$ generator technology. It is basically an alumina functionalized by sulfate to adsorb much more ^{188}W than the usual alumina used for the adsorbent. The ^{188}W is produced by 2n reactions of ^{186}W. Though the neutron capture cross section of ^{186}W is large, the 2n reactions give very low specific activity of ^{188}W even at the highest neutron flux available over the world, which resembles the case of $(n,\gamma)^{99}Mo$. The similar chemical characteristics of Mo and W allow a high adsorbing of the Mo in the ALSUL as well. Experiments showed that adsorbing at least 0.2 g-Mo/g-ALSUL is possible, which is comparable to the PZC. An advantage of the ALSUL is that the process to make the ^{99m}Tc generator column is the same for the conventional alumina column using fission Mo, and therefore the conventional ^{99m}Tc generator loading facility can be used without significant modification.

3. Motive of feasibility study on $(n,\gamma)^{99}Mo$ production at HANARO

As explained in Section 2, backup reactors from nearby countries are needed for the production of $(n,\gamma)^{99}Mo$ at HANARO for stable availability of ^{99m}Tc in Korea. At the 2008 KAERI-JAEA Joint Seminar on Advanced Irradiation and PIE Technology, held at KAERI in November, JAEA expressed a strong intention to produce $(n,\gamma)^{99}Mo$ at JMTR [2,3]. The construction of CARR (China Advanced Research Reactor) was almost completed as well. Therefore, it was expected that backup reactors would be available.

However, if the role of HANARO is limited to emergency supply only as explained in Section 2, it is not only inefficient for managing relevant manpower and equipment but also impractical to establish a robust backup supply scheme because its demand is not routine but accidental. Therefore, maintaining a sizable supply with self sustaining capability is desired. As the world supply is instable and the future seems not to be better, such supply is demanding and can be fully justified. But it is possible when the $(n,\gamma)^{99}Mo$ has a certain capability to compete with the fission Mo.

In Korea, all ^{99m}Tc is normally obtained from portable ^{99m}Tc generators. It is already proven that the supply of ^{99m}Tc solution from HANARO cannot compete with ^{99m}Tc generators for the normal supply. Therefore, while the production of $(n,\gamma)^{99}Mo$ is only for the supply of ^{99m}Tc solution, it is always limited to the emergency supply only. There would be two ways to achieve the sizable supply with self sustaining capability. First is developing a new competitive ^{99m}Tc generator technology using a high Mo adsorbent, and second is establishing a new mass supply scheme of ^{99m}Tc nuclear medicines from the $(n,\gamma)^{99}Mo$ rather than the supply of ^{99m}Tc solution. The first option is attractive because the current ^{99m}Tc generator loading facility can be utilized and the current domestic scheme from ^{99}Mo supply to use of ^{99m}Tc can be maintained without significant change. But it requires a new generator technology competitive. It is expected that ALSUL has such potential. The second option may be possible by refining and checking the current ^{99m}Tc solution extraction technology from (n,γ)^{99}Mo, but disturbs the current domestic scheme significantly.

It is also supposed that the most important factor for the viability of the (n,γ)^{99}Mo is the success of a new high adsorbent. If any one of the high Mo adsorbents including the ALSUL and the PZC is successful to make competitive ^{99m}Tc generators, it will encourage many high flux reactors to produce the (n,γ)^{99}Mo. The (n,γ)^{99}Mo is much easier than the fission Mo not only in the post processing but also for the target irradiation. Therefore, many high flux reactors over the world may be available for the (n,γ)^{99}Mo production. The supply scheme of the (n,γ)^{99}Mo, if it is realized, must be different from the fission Mo, because the specific activity of the (n,γ)^{99}Mo is not high enough even with the high adsorbent. The shortage in the low specific activity can be alleviated by utilizing high neutron flux and enriched ^{98}Mo targets, but minimization of ^{99}Mo decay time is still necessary, which can be achieved by limiting its supply to short distance. Then, a regional supply by a group of high flux reactors in each region can be an effective and reliable way.

In this region, potential reactors for the cross backup supply are JMTR and JRR-3 in Japan, CARR and HFETR (High Flux Engineering Test Reactor) in China, and RSG-GAS in Indonesia. The JMTR is under refurbishment and intends to produce (n,γ)^{99}Mo when it resumes operation in 2011. The JRR-3 may be utilized together for the backup supply. The HFETR in NPIC (Nuclear Power Institute of China), Chengdu, has been supplying (n,γ)^{99}Mo to southern China together with MJTR (Min Jiang Test Reactor) in the same site. Its isolated location requires relatively long time for the transportation of ^{99}Mo, but the very high neutron flux of the reactor may compensate the extended decay for the transportation. HWRR (Heavy Water Research Reactor) and SPR (Swimming Pool Reactor) in CIAE (China Institute of Atomic Energy), Beijing, had been supplying fission Mo to northern China. The HWRR was permanently shut down at the end of 2007 and the CARR will begin operation soon. Since they have been operating a fission Mo processing facility, it is not certain whether they will join the regional backup supply of (n,γ)^{99}Mo. But at least they may supply fission Mo when other reactors in the region are not available. The situation of RSG-GAS in Indonesia would be similar as the CARR except relatively long distance.

In order to be backed up, HANARO may also have cross backup capability, which means the peak ^{99}Mo production capability of HANARO should be much larger than its domestic supply.

The potential of (n,γ)^{99}Mo production at HANARO was studied from above view points.

4. Selection of irradiation holes and targets for analysis

Figure 1 shows a horizontal model of HANARO core and irradiation holes for MCNP [7] calculations. Three irradiation holes – IR2, OR3 and IP15 are selected for a sensitivity analysis. Since they have distinctive neutron spectra very different each other, a good guide line for the search of an optimum irradiation from a neutronic viewpoint is expected. For the selection of an irradiation hole for the actual $(n,\gamma)^{99}Mo$ production, other factors should be taken into account.

The IR2 is one of hexagonal flux trap in the core, in which all fast, epithermal and thermal neutron fluxes are high. The OR3 is a circular hole of 60 mm inner diameter located in a heavy water reflector region near the core boundary, in which epithermal and thermal neutron fluxes are high. The IP15 is the same circular hole in the heavy water reflector region but farther than the OR3 from the core boundary, in which only thermal neutron flux is high.

All the same horizontal target dimensions in the three holes are assumed as current radioisotope production capsules for ^{131}I and ^{192}Ir. A 28 mm diameter and 600 mm length Mo target encapsulated in a double walled aluminum capsule is located at horizontally and vertically central position in the hole. Actual length of each capsule is not so long and multiple capsules are irradiated at a hole simultaneously. But the 600 mm is assumed for the convenience' sake in calculation and data analysis, while an active fuel length is 700 mm. Horizontal dimensions of OR3 and IP15 with targets are shown in Figure 2. For the case of IR2, dimensions only outside of the aluminum pipe are different as shown in Figure 1.

Each target region is divided into inner and outer sections at a half radius of the target, and then into left and right sections each, to investigate effects from the neutron self shielding and of distance from the core center. While the left hand side is closer to the core center at IR2 and IP15, the right hand side is closer to the core center at OR3.

Calculations were done for seven different artificial targets; 1) natural MoO_3 powder, 2) natural MoO_3 pellet, 3) enriched $^{98}MoO_3$ pellet, 4) homogeneous mixture of enriched ^{98}Mo metal (60% volume fraction) and water, 5) enriched ^{98}Mo metal shots (almost 60% volume fraction) of 2 mm diameter close packed in the water, 6) enriched ^{98}Mo metal at 300 K, and 7) enriched ^{98}Mo metal at 1000 K. Targets are assumed to have 1.45, 3.5 and 10.2 g/cm^3 density for MoO_3 powder, MoO_3 pellet and Mo metal, respectively, and 24.13% and 98.5% ^{98}Mo for natural and enriched ^{98}Mo, respectively.

The 1.45 g/cm^3 MoO_3 powder is chosen based on the experience at HANARO. JAEA [2] had made 3.25 ~ 3.4 g/cm^3 MoO_3 pellets and expected further increase of the pellet density. Therefore, the 3.5 g/cm^3 is chosen for the MoO_3 pellet. After then, the JAEA [6] succeeded in 4.45 g/cc MoO_3 pellet manufacture, which is about 95 % of theoretical density (4.69 g/cm^3). The 98.5% enriched ^{98}Mo is chosen based on experience at HANARO (see Section 2). The mixtures of Mo metal and water are chosen to check the effect of water to the specific activity of ^{99}Mo as well as a potential way increasing the ^{98}Mo loading. The homogeneous mixture is a simplified model for the metal shots close packed in the water. Calculations confirmed that the homogeneous mixture model gives almost the same results as the heterogeneous model. The 1,000 K metal temperature in addition to 300 K is chosen to find out the effect of Doppler broadening in ^{99}Mo production, which is well known in reactor physics that the Doppler broadening increases the resonance absorption.

5. Analysis of calculated results

Figure 3 shows variations of total ^{98}Mo(n,γ) reaction rates per g-Mo and g-^{98}Mo depending on the ^{98}Mo loading density at respective irradiation holes. Data points at two lowest ^{98}Mo loading densities at the left hand side in the figure are for natural Mo, and others are for enriched ^{98}Mo. The trend of reaction rates per g-^{98}Mo indicates neutron flux depression and self shielding, whereas the trend of reaction rates per g-Mo is identical with that of ^{99}Mo specific activity. If there is neither change in neutron flux nor spectrum by different targets, the reaction rates per g-^{98}Mo must be all the same. However, they are decreasing due to neutron self shielding as the ^{98}Mo loading increases, with high sensitivity in the hard neutron spectrum and for the density of natural Mo targets.

Reasons of trends shown in Figure 3 are easily understood from quite different neutron capture cross sections between natural Mo and ^{98}Mo as shown in Table 1 and Figure 4. Neutron capture cross sections of natural Mo are about 20 and 4 times of ^{98}Mo in thermal and epithermal energy ranges, respectively. Therefore, the neutron self shielding in and the flux depression from the natural Mo targets are basically larger than the enriched ^{98}Mo targets. In the epithermal energy range, the ^{98}Mo has many narrow resonance absorption peaks and the resonance self shielding will be basically determined by the ^{98}Mo loading. For the case of fast neutrons, cross sections for both of natural Mo and ^{98}Mo are very small. Therefore, the portion of fast neutron reaction rate of ^{98}Mo in its total reaction rate is small and its effective cross section is not sensitive, regardless of target.

From Figure 4, we can also imagine that the portion of epithermal neutron reaction rate for ^{99}Mo production by Mo targets will be much larger than that by ^{235}U. It also indicates that a hard neutron spectrum with high epithermal neutron flux will be good for the (n,γ)^{99}Mo production, whereas a well thermalized neutron spectrum will be enough for the fission Mo production

In order to understand the effect of neutron self shielding quantitatively, respective reaction rates for thermal, epithermal and fast neutrons are investigated. Neutron energy boundaries dividing three neutron energy groups are 0.625 eV and 0.1 MeV in this study. The ^{98}Mo has $1/v$ cross sections up to about 1 eV and its sharp resonance peaks are found up to 32 keV in the ENDF/B-VII neutron cross section library as shown in Figure 4. In the JEFF-3.1 library, however, its sharp resonances are found up to 12 keV only. Therefore, the ^{98}Mo may actually have many sharp resonances above 32 keV as well but may not be evaluated in the ENDF/B-VII yet. Even many sharp resonances above 32 keV are represented by average values as in Figure 4, since the cross sections are in decreasing trend at above 32 keV and the cross section around 32 keV is much lower than those at the lower energy, its impact to the resonance reaction rates must be small. Therefore, it can be assumed that while the thermal neutron reaction rate below 0.625 eV and the fast neutron reaction rate above 0.1 MeV are free of the sharp resonance absorption, the epithermal reaction rate is almost due to the resonance integral.

Figure 5 shows thermal neutron flux and reaction rates per g-Mo depending on the ^{98}Mo loading density at respective irradiation holes. For the cases of natural Mo targets, thermal neutron fluxes are rather sensitive to Mo loading densities due to relatively high thermal neutron capture cross sections. Fluxes with enriched Mo targets are slightly higher due to much lower thermal neutron capture cross sections of the enriched Mo. ^{98}Mo cross sections are low enough not to reduce the flux noticeably even at very high loading density of pure Mo metal target. When the Mo metal shots are mixed with water, the thermal neutron flux and reaction rate are notably large in the in-core irradiation hole IR2. It is because the water slows down fast and epithermal neutrons.

The thermal neutron reactions per g-^{98}Mo are not depicted in the figure because they are almost proportional to the thermal neutron flux. It means that effective thermal neutron cross sections are not sensitive to the ^{98}Mo loading density. Effective thermal neutron cross sections averaged over different targets are 0.0998 b, 0.1082 b and 0.1098 b for IR2, OR3 and IP15, respectively, which are a little smaller than the Maxwell average cross section 0.1153 b in Table 1. The trend clearly shows the smaller effective thermal neutron cross section for the harder thermal neutron spectrum, but very little difference from different targets.

For the case of epithermal neutrons, the neutron fluxes are little influenced from different targets as shown in Figure 6. But their reaction rates per g-^{98}Mo are very sensitive to the ^{98}Mo loading density, which consequently gives very different effective neutron cross sections. The effective epithermal neutron cross sections versus logarithm of ^{98}Mo loading density are plotted in Figure 7 for inner and outer parts of targets separately. Since very narrow resonance reactions give rather poor statistics in Monte Carlo calculations, data divergence at low ^{98}Mo loading becomes large, but we can say that the effective epithermal neutron cross sections have certain correlation with the ^{98}Mo loading, which is almost independent of the irradiation holes. Exceptions are mixed metal shots mixed with water and metal targets at 1000 K. They are larger than the correlated values as expected. The water mixed with Mo metal shots scatters epithermal neutrons causing some of them fall in the flux wells made by resonance self shielding, and consequently reduces the resonance self shielding effect and increases the effective cross sections. When the Mo metal temperature is high, resonances are broadened by Doppler effect, and the effective cross sections become larger.

Wells in neutron spectrum at the resonance peaks become deeper at the inner parts of targets. Therefore, the resonance self shielding effect is larger in the inner parts, and cross sections for inner parts are lower than outer parts. The effective cross sections versus logarithm of the ^{98}Mo loading are almost on straight lines. The ^{98}Mo loading can be expressed as linear loading in the cylindrical rod type targets, say g-^{98}Mo per cm, as well. When average values of effective epithermal neutron cross sections over three irradiation positions are least square fitted to the ^{98}Mo linear loading without two exceptional cases aforementioned, they become Figure 8. Table 2 lists coefficients fitted.

The effects of 40% volume fraction water mixed with Mo metal shots can be obtained by comparing with fitted values. It increases effective cross sections by about 32%, 22% and 24% for inner, outer and overall targets, respectively. Since MoO_3 targets have three oxygen atoms per Mo, it may contribute to reducing the resonance self shielding but, when they are compared with Mo metal, actually it is not distinguished in Figures 7 and 8. Its reason is explained by Figure 9 which compares elastic scattering cross sections for nuclides composing targets. From the figure, it is clear that the oxygen cannot contribute to the total scattering so much due to its low scattering cross sections at high neutron energy compared to Mo.

The effect of Doppler broadening can be obtained from data at 300 K and 1,000 K. It increases the effective cross sections by about 15% regardless of inner or outer parts of targets.

Effective cross sections without any resonance self shielding (infinitely diluted medium) can be estimated from the resonance integral 6.553 b for ^{98}Mo in Table 1. The resonance integral is defined as following equation;

$$I = \int_{E_1}^{E_2} \frac{\sigma(E)}{E} dE \tag{1}$$

where, I is resonance integral, and E_1 and E_2 are lower and upper energy boundaries of resonance region. Above equation is an ideal case that the neutron spectrum is $1/E$ without any resonance shelf shielding. Meanwhile, the epithermal neutron flux and reaction rate can be approximated by integrated values of $1/E$ spectrum, over epithermal neutron energy range.

$$\phi = \int_{E_1}^{E_2} \frac{\varphi}{E} dE = \varphi \ln(\frac{E_2}{E_1}) \tag{2}$$

where, ϕ is epithermal neutron flux and φ is magnitude of $1/E$ spectrum. Then, the epithermal neutron reaction rate for the $1/E$ spectrum is expressed as following;

$$1/E \text{ Reaction Rate} = \int_{E_1}^{E_2} \frac{\varphi\sigma(E)}{E} dE = \varphi I = \frac{\phi I}{\ln(\frac{E_2}{E_1})} = \sigma_{epi}\phi \tag{3}$$

where, $\sigma_{epi} = \frac{I}{\ln(\frac{E_2}{E_1})}$. (4)

In this calculations, E_1 = 0.625 eV and E_2 = 0.1 MeV. Therefore, σ_{epi} = 0.08345 I = 0.5469 b. When this value is compared with values in Figure 7, we can find that the resonance self shielding effects are significant. It is also found that the effective epithermal neutron cross section is larger than the effective thermal neutron cross section, except the inner part of enriched Mo metal target. Therefore, it can be said that high epithermal neutron flux is effective for the ^{99}Mo production in general, but the resonance self shielding effect is also very large.

For the case of fast neutron reactions, the effective cross sections are around 0.033 b regardless of irradiation positions and targets. The value is larger than the fission spectrum average value 0.02653 b in Table 1. It is because actual fast neutron spectra above 0.1 MeV are softer than the fission spectrum.

6. Capability of HANARO for the supply of $(n,\gamma)^{99}$Mo

The supply of $(n,\gamma)^{99}$Mo is possible when the product satisfies users' requirements. As explained in Section 3, one of objectives of the calculation is to check whether the sizable production of ^{99}Mo for portable ^{99m}Tc generators is possible assuming that the ALSUL is fully verified. Then, first of all, ^{99}Mo specific activity has to be high to make competitive ^{99m}Tc generators by the ALSUL.

In Korea, activity of a generator calibrated at 12:00 Wednesday of the week under use is in 0.2 ~ 1.5 Ci range. When these generators are made of $(n,\gamma)^{99}$Mo, the size of generator including shielding has to be similar with usual generators using fission Mo. Otherwise, the $(n,\gamma)^{99}$Mo generators cannot compete in the market. In USA, the price portion of ^{99}Mo solution is estimated about 38% in generators, 25% in the dose to patients, and less than 1% in the ^{99m}Tc imaging processes [7]. The portion becomes smaller as generator activity is lower because of a fixed cost independent on the generator activity. Generators in Korea are all portable generators used in hospitals, of which activities are much lower than jumbo generators used by radiopharmacies. Then, the portion of ^{99}Mo solution in Korea would be lower than USA. Therefore, even though the cost for the production of ^{99}Mo solution is significantly reduced, if it increases cost for the next processes, its impact has to be carefully examined.

The most critical factor influencing the cost in the next processes is volume of the column. A larger column for a same ^{99}Mo activity not only requires heavier and larger shielding, but also gives lower volumetric ^{99m}Tc activity. It is assumed that a ^{99m}Tc generator containing 3 g ALSUL column can be used almost the same way as the conventional portable ^{99m}Tc generators using fission Mo. The column is still larger than conventional columns, but it is supposed that its impact is small enough to be sufficiently compensated by the lower cost of $(n,\gamma)^{99}$Mo. It is also assumed that the maximum amount of Mo adsorbed by 1 g-ALSUL is 0.2 g. Therefore, 0.6 g is the maximum amount of Mo per ^{99m}Tc generator.

Since loading and unloading process of $(n,\gamma)^{99}$Mo targets are very simple, one hour is assumed for the replacement of targets. Therefore, target irradiation time is 6 days and 23 hours. As the post processing of irradiated targets is also simple, it is assumed that irradiation up to 09:00 Sunday would be possible for the delivery of ^{99m}Tc generators to the hospitals in the Sunday evening for their use from Monday morning. Then decay time of Mo to 12:00 Wednesday from end of irradiation is 3 days and 3 hours.

The specific activity of ^{99}Mo at the time of calibration is expressed as;

$$A_S = R(1-e^{-\lambda T_I})e^{-\lambda T_D} \tag{5}$$

where A_S = specific activity of ^{99}Mo in Bq/g-Mo, R = specific ^{98}Mo(n,γ) reaction rate in /s/g-Mo, λ = ^{99}Mo decay constant in /h, T_I = irradiation time, and T_D = decay time after irradiation in h. Specific activities of ^{99}Mo and its production rates per 1 cm target calibrated at 12:00 Wednesday are listed in Table 3. In order to make the highest activity generator in Korea (1.5 Ci) from 0.6 g Mo, the specific activity has to be higher than 2.5 Ci/g-Mo. The table shows that it is possible when enriched ^{98}Mo is irradiated at IR2 and OR3.

In actual situation, however, while the IP15 is readily available for the ^{99}Mo production, IR2 and OR3 are not. Since the IP15 is located outside of forced convection flow region of primary coolant, targets are routinely loaded and unloaded for the isotope production during power operation. But the specific activity of ^{99}Mo from the IP15 is not high to make 1.5 Ci generators. This hole can be utilized for the ^{99m}Tc solution supply for which the high ^{99}Mo activity is not mandatory, or for the supply of low activity generators below 0.8 Ci using enriched ^{98}Mo targets at the initial stage of (n,γ) ^{99}Mo generator supply.

Use of IR2 for the ^{99}Mo production is possible only when the reactor is operated weekly mode. On power loading and unloading of targets using a hydraulic rabbit system are not practical in this hole due to interference with refueling and impact to the core. The on-power loading and unloading at an OR hole will be possible if a hydraulic rabbit system is installed. Therefore, the capability of HANARO to supply (n,γ)^{99}Mo is discussed for the case that the OR3 hole is ready for the (n,γ)^{99}Mo production with a hydraulic rabbit system.

As shown in Table 3, the production capability of OR3 is 39.4 Ci/cm when 3.5 g/cm^3 enriched ^{98}Mo pellets are irradiated. As analyzed in the Appendix, weekly demand in Korea is about 100 Ci/w calibrated at 12:00 Wednesday. This demand can be satisfied by irradiating only a target a little longer than 2 cm.

The regional demand analyzed in the Appendix is about 1,500 Ci/w. The backup supply to other countries may require longer time for transportation than domestic supply. One additional day would be enough for the transportation. The ^{99}Mo decays to 77.7% in a day. Even the decay is taken into account, the hole has capability to produce 1,500 Ci/w. The production capability can be increased by using 4.45 g/cm^3 pellet instead of 3.5 g/cm^3 and there is some room to increase the target diameter as discussed in the next Section. As neutron flux at other OR holes are similar with the OR3, it can be said that an OR hole can produce a large amount of (n,γ)^{99}Mo sufficient to satisfy the regional demand. It is possible by irradiating enriched $^{98}MoO_3$ pellets. Therefore, irradiation of Mo metal targets, which requires additional study for their post processing, is not necessary.

7. Potential hydraulic rabbit system at HANARO for the $(n,\gamma)^{99}Mo$ production

Reactor vessel of HANARO is open to the pool at the top. Therefore, the hydraulic rabbit system at an OR hole do not have pressure boundary related to the reactor safety and the maximum pressure applied to the irradiation part of the rabbit system is within a few bars. It allows use of aluminum for the parts under neutron irradiation. Potential horizontal dimensions of the rabbit irradiation part are shown in Figure 9. The water thickness both at the surface of rabbits and between inner and outer pipes is assumed as 2 mm. The length of narrow water gap between the inner and outer pipes is limited to about 1 m. The OR is a 6 cm diameter hole in the 1.2 m height heavy water tank. Above the heavy water tank, the diameter of pipes can be enlarged to have a sufficient water gap thickness. The 1.5 mm He gap between inner and outer walls of current RI capsule is reduced to 0.25 mm in the rabbit. This gap size must be minimized while the inner capsule can be inserted in the outer rabbit. The larger gap gives no advantage but increases the temperature of target. If the pellet is sufficiently stable in the water, single containment rabbit can also be considered.

The pellet diameter in Figure 9 is 32 mm, which is 4 mm larger than the current target. As the pellet diameter becomes larger, the pellet center line temperature increases almost in proportion to a square of the diameter.

It seems that the pellet temperature won't be higher than MoO_3 sublimation temperature 750 °C under the gamma heating of the HANARO OR hole even though the pellet diameter is maximized. When a very high temperature is predicted, a cooling hole at the center of rabbit effectively reduces the temperature. If the pellet diameter can be 32 mm and 4.45 g/cm^3 enriched $^{98}MoO_3$ is used, the production capability of an OR hole can reach to about 3,000 Ci/w which is about twice of estimated regional demand.

A space beneath the rabbits is not enough to install a floater for the replacement of rabbits when they are unloaded. But a floater for a hydraulic damper to reduce mechanical impact of rabbits when they are loaded may be installed.

8. Concluding remarks

After the feasibility study on $(n,\gamma)^{99}Mo$ production at HANARO was published by a KAERI report [4], worldwide supply of ^{99}Mo became worse. On 16 February 2010, Korean Government decided to operate HANARO for the ^{99m}Tc supply with the highest priority, if shortage of ^{99}Mo in Korea occurs. On 12 March 2010, FNCA National Coordinators' meeting decided to discuss establishment of a regional radioisotope network. The need for early available alternative ^{99}Mo became stronger. Previous study indicated that the $(n,\gamma)^{99}Mo$ has potential to be an alternative mass ^{99}Mo available earlier than any other options. Calculation for HANARO indicated that each high flux reactor in the region may have large $(n,\gamma)^{99}$Mo production capability as well. Therefore, if high flux reactors in the region collaborate in a concerted mode, a stable supply scheme of $(n,\gamma)^{99}$Mo can be established.

It becomes practical when radioisotope industry of each country accepts the use of $(n,\gamma)^{99}$Mo for a meaningful portion of national demand. Though $(n,\gamma)^{99}$Mo has many advantages compared to fission Mo, its use over the world is almost negligible because it is less convenient to users than fission Mo. Efforts have been made to improve its convenience in use especially by developing high Mo adsorbents such as Zr gel generator, PZC and ALSUL, but it cannot be better than the use of fission Mo. As the portion of ^{99}Mo in the payment of patients diagnosed by using ^{99m}Tc is almost negligible, users prefer the convenient fission Mo rather than less expensive $(n,\gamma)^{99}$Mo. However, it is well known that the fission Mo has risk of nuclear proliferation and release of fission products, and produces much radwaste. It would not be known so well that the fission Mo has an inherent risk in supply due to global supply from a few reactors [7], as well. The $(n,\gamma)^{99}$Mo has virtually no such problems.

The current world situation relevant to ^{99}Mo may be figured as patients suffering from diseases caused by fast food. As healthy food is recommended to the patients, $(n,\gamma)^{99}$Mo can be recommended to them for risk free ^{99}Mo. If they earnestly wish the stable availability of ^{99}Mo for patients and are willing to work more than before, the $(n,\gamma)^{99}$Mo will be available soon. It will reduce not only above risk but also much dirty jobs dealing with HEU and highly radioactive materials and waste. Instead, it may create new jobs with almost negligible impact to payment of patients. Meanwhile, close collaboration among high flux reactors is essential. This report is made as a result of collaboration between HANARO and JMTR. We need collaboration with other reactors in the region, as well.

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Table1. Neutron capture cross sections of natural Mo and ^{98}Mo [13]

unit: b

Group	Natural Mo	^{98}Mo
Maxwell Average	2.288	0.1153
Resonance Integral	25.67	6.553
Fission Spectrum Average	0.03163	0.02653

Table 2. Fitted coefficients for effective epithermal neutron ^{98}Mo(n,γ) cross section averaged over 60 cm target length

Region	A	b	
Inner	-0.06808	0.3536	$\sigma = a\ln(\rho') + b$
Outer	-0.07058	0.3972	σ = Effective epithermal ^{98}Mo(n,γ) cross section (b)
Over all	-0.06996	0.3863	ρ' = Linear ^{98}Mo loading (g/cm)

Table 3. ^{99}Mo specific activity and production rate calibrated at 12:00 Wednesday

Axially averaged specific activity for 60 cm (Ci/g-Mo) / Production rate (Ci/cm)

Target (density: g/cm^3)	^{98}Mo Loading (g/cm^3)	IR2	OR3	IP15
Natural Mo powder (1.45)	0.233	1.56/9.2	0.87/5.1	0.40/2.3
Natural Mo pellet (3.5)	0.563	1.34/19.0	0.78/11.1	0.34/4.8
Enriched Mo pellet (3.5)	2.288	4.19/59.6	2.77/39.4	1.38/19.6
40% water + 60% enriched Mo metal	5.998	3.95/148.7	2.59/97.7	1.27/47.8
Enriched Mo metal (10.2)	9.996	3.00/188.5	2.25/141.4	1.17/73.7
Enriched Mo metal 1000K	9.996	3.17/199.0	2.34/147.1	1.19/75.0
Ratio of peak 20cm to average 60cm	-	1.22	1.16	1.16

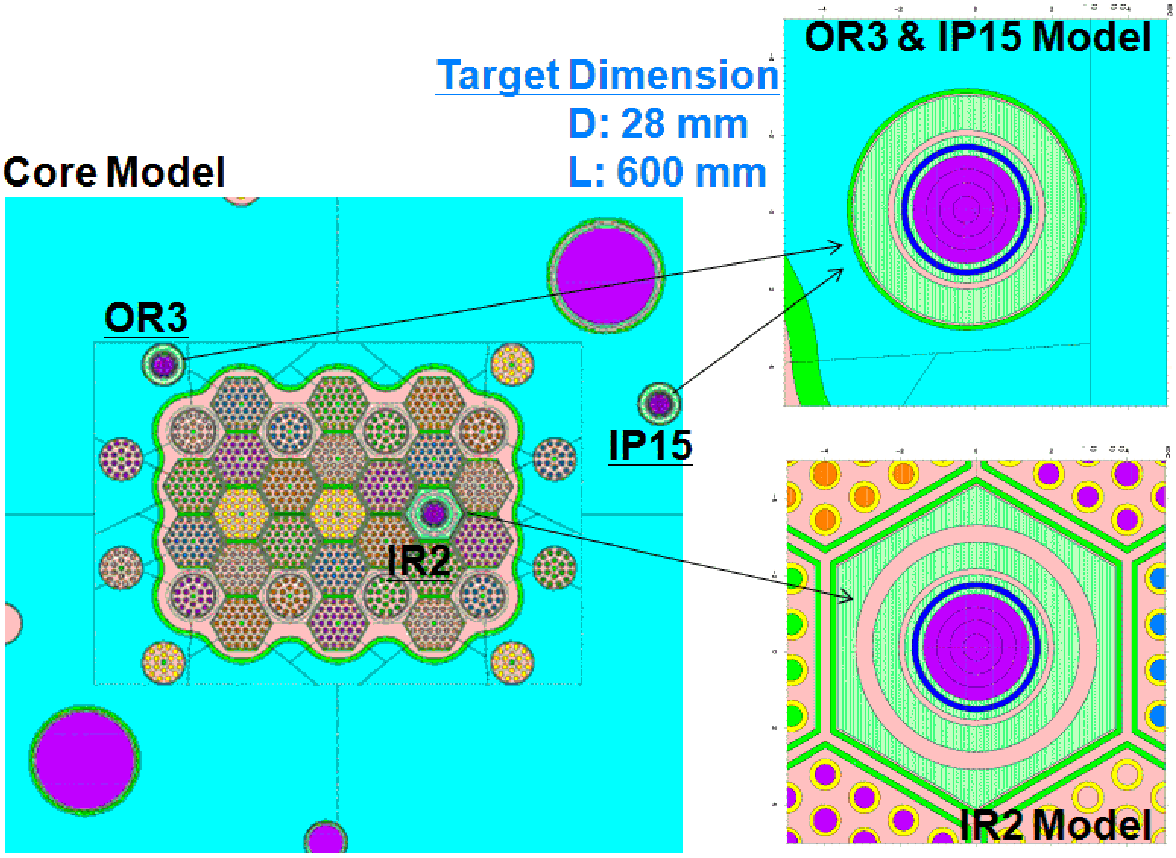

Figure 1. Horizontal MCNP model of HANARO core and irradiation holes

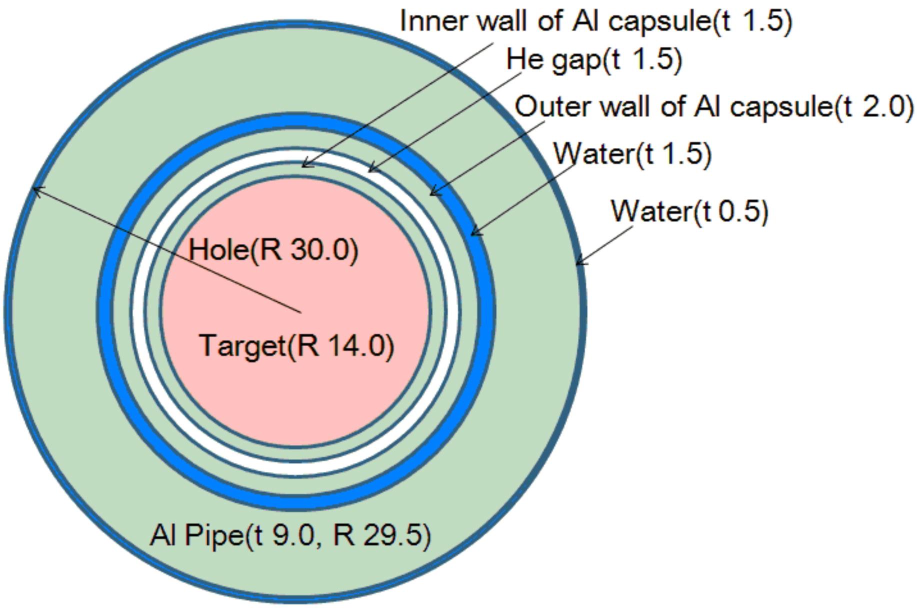

Figure 2. Horizontal dimensions of OR3 and IP15 holes for RI production (unit: mm)

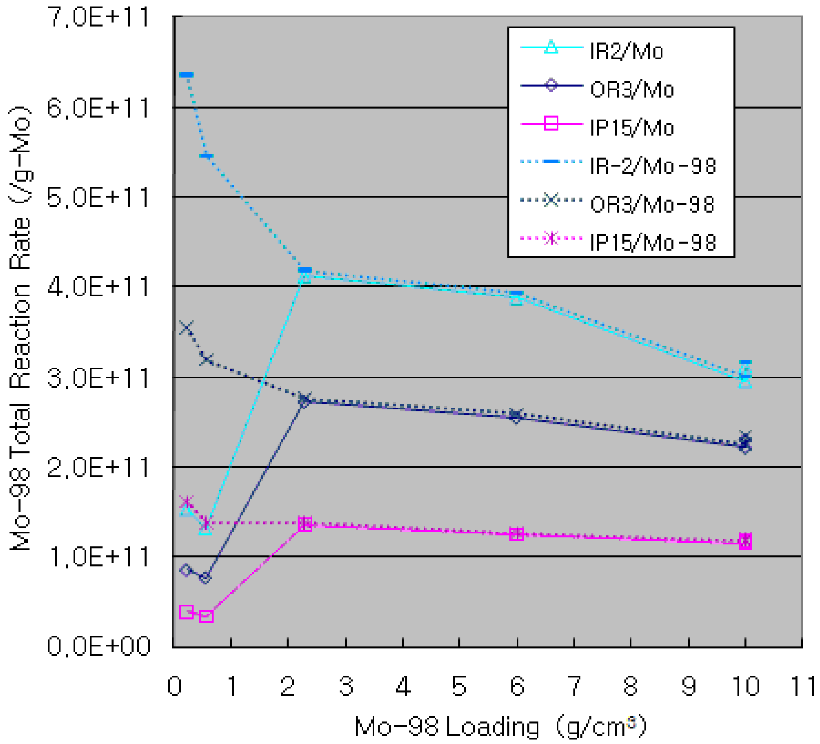

Figure 3. Total ^{98}Mo(n,γ) reaction rate per g-Mo and per g-^{98}Mo averaged over 60 cm target length

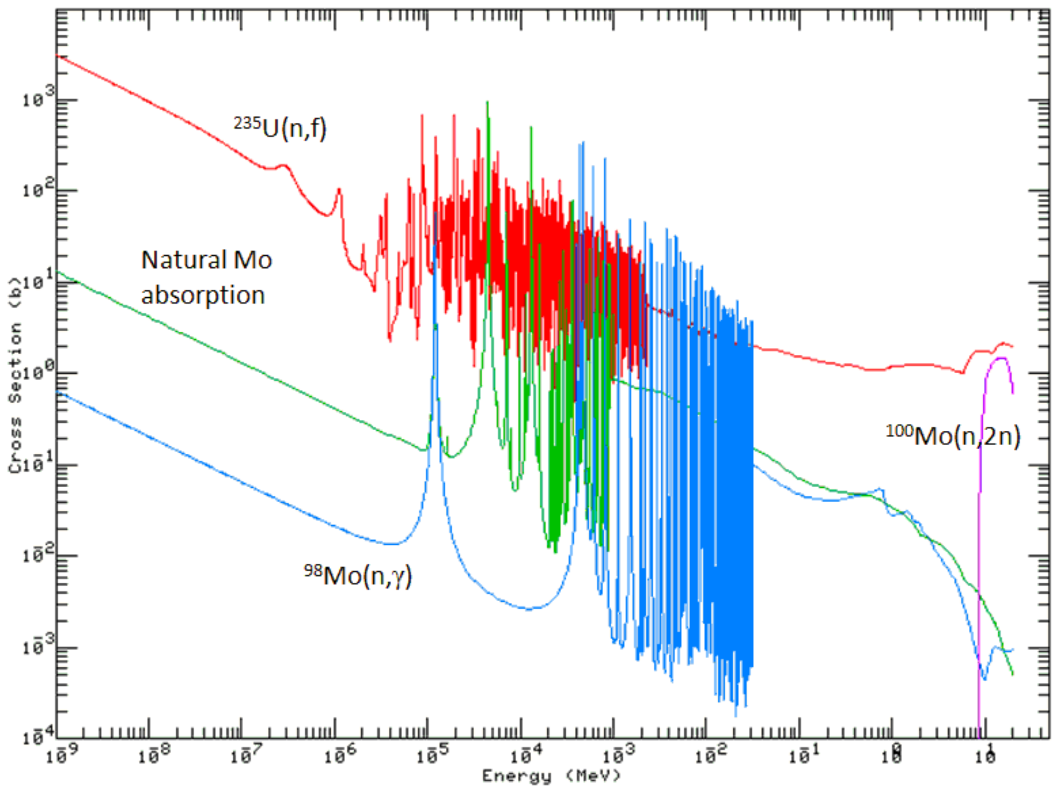

Figure 4. Cross sections of nuclides for the ^{99}Mo production [14]

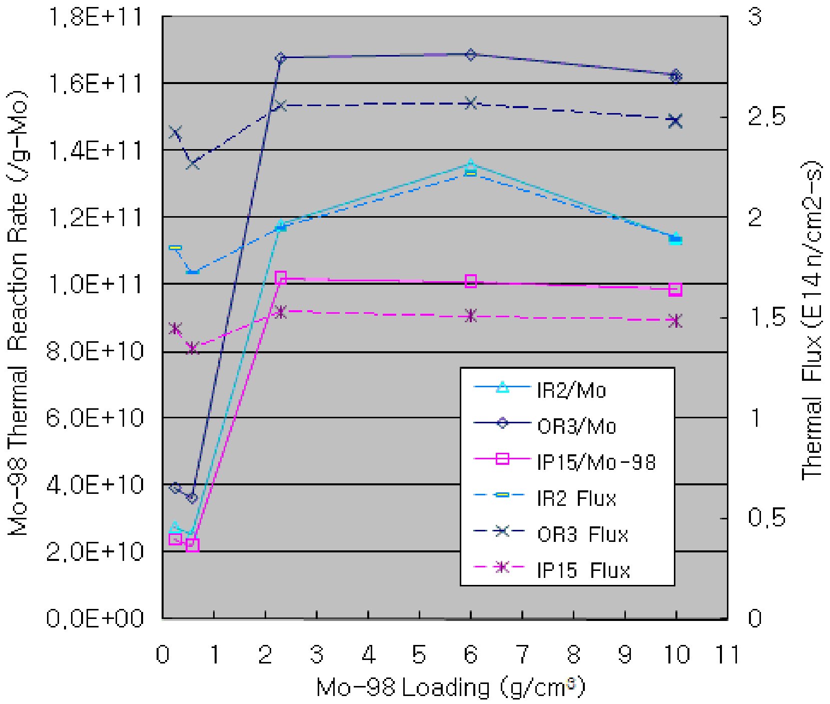

Figure 5. Thermal neutron flux and ^{98}Mo(n,γ) reaction rate per g-Mo averaged over 60 cm target length

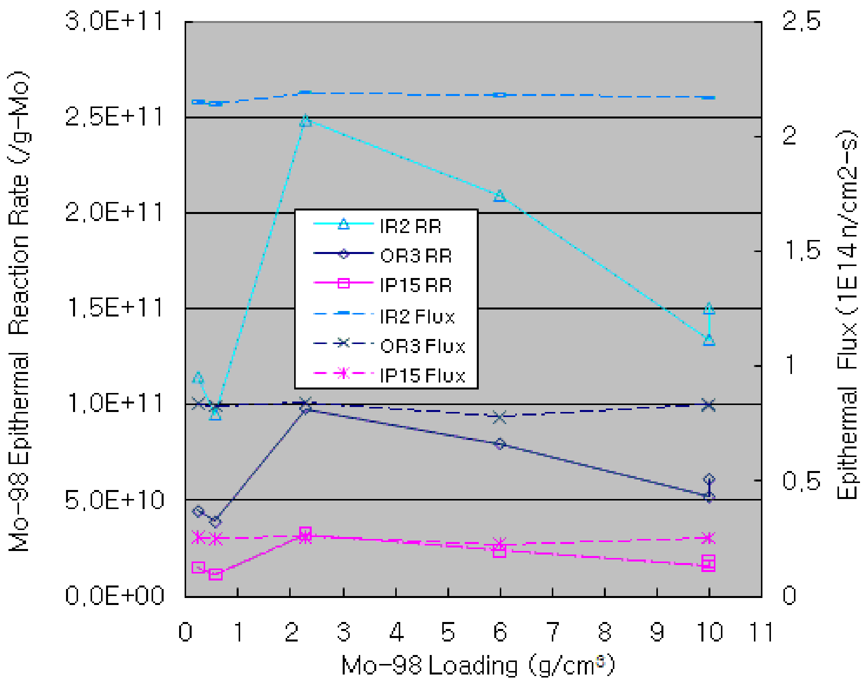

Figure 6. Epithermal neutron ^{98}Mo(n,γ) reaction rate per g-Mo averaged over 60 cm target length

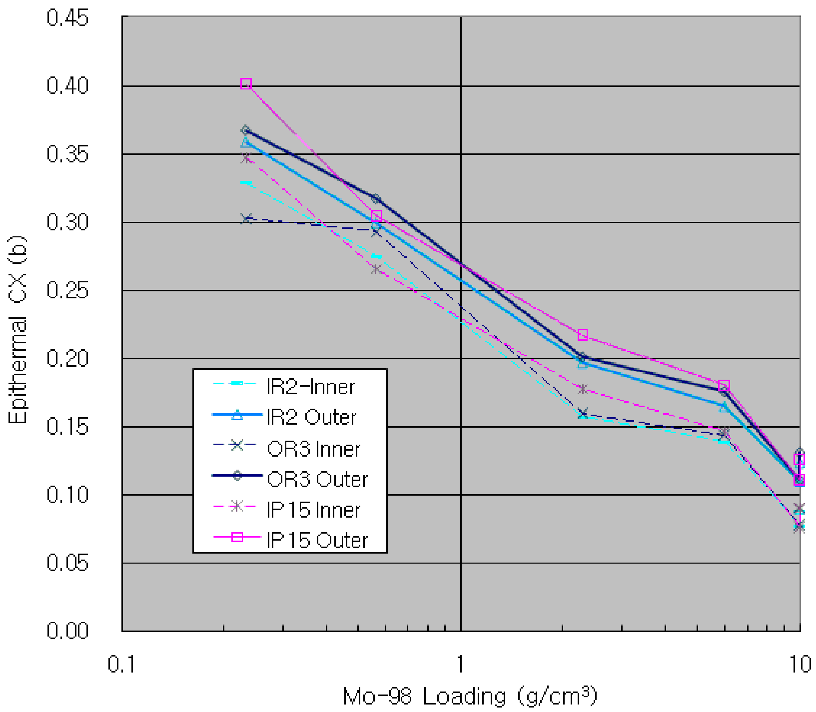

Figure 7. Effective epithermal neutron ^{98}Mo(n,γ) cross section averaged over 60 cm target length

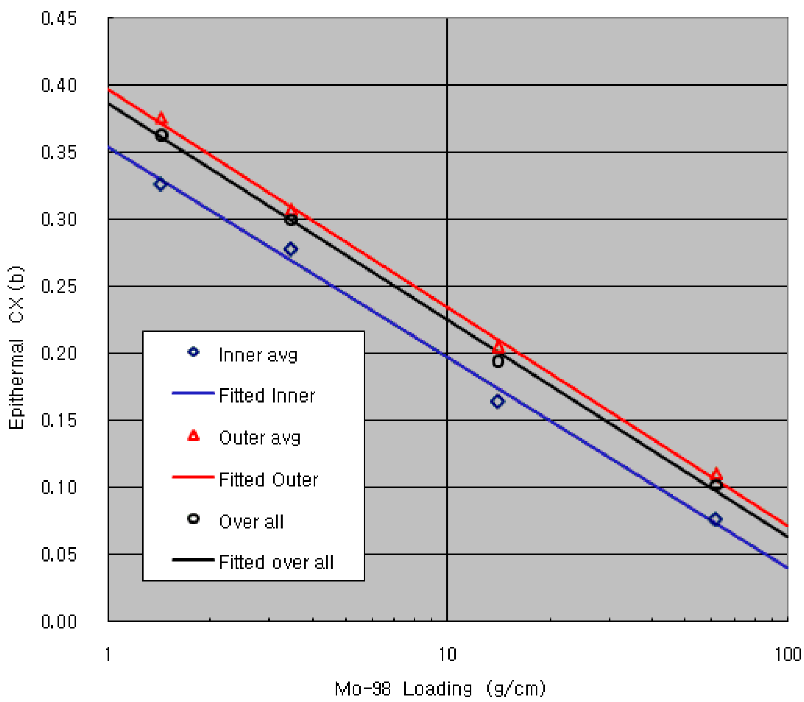

Figure 8. Fitted effective epithermal neutron ^{98}Mo(n,γ) cross section for linear loading of ^{98}Mo averaged over 60 cm target length

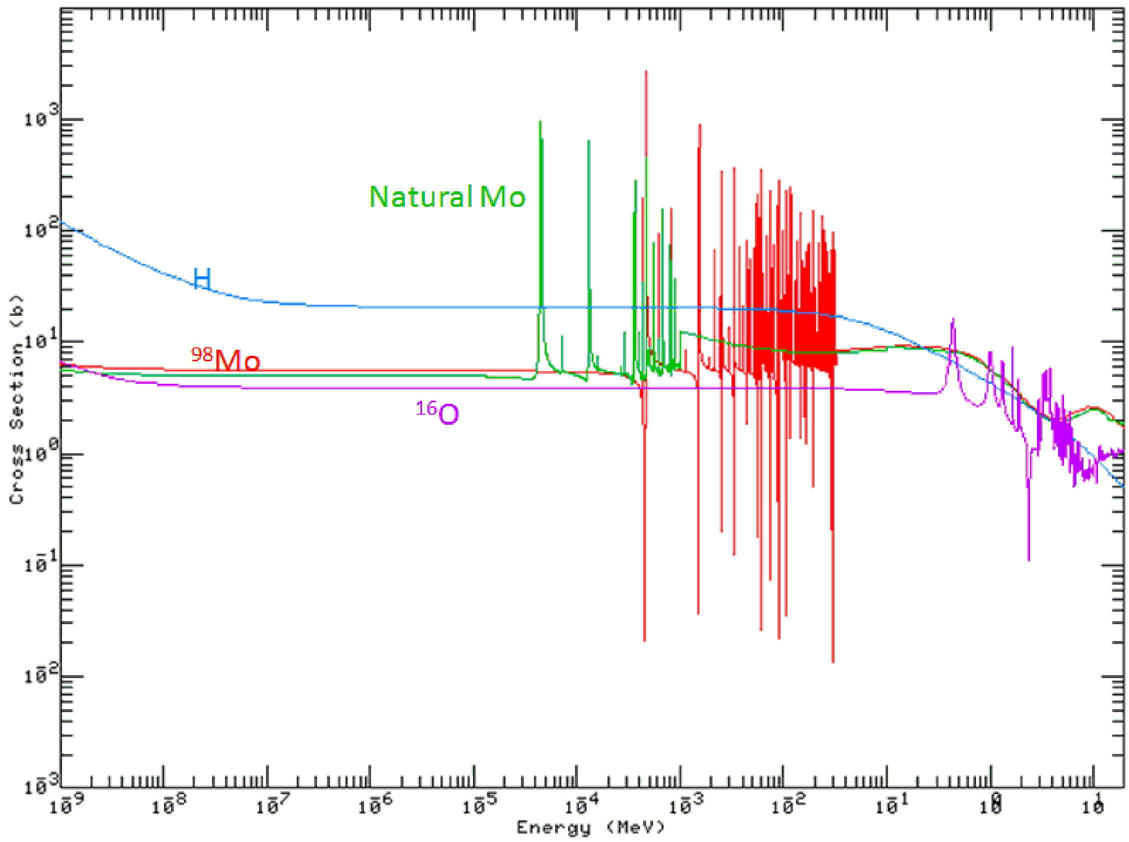

Figure 9. Elastic scattering cross sections of materials of $(n,\gamma)^{99}$Mo target

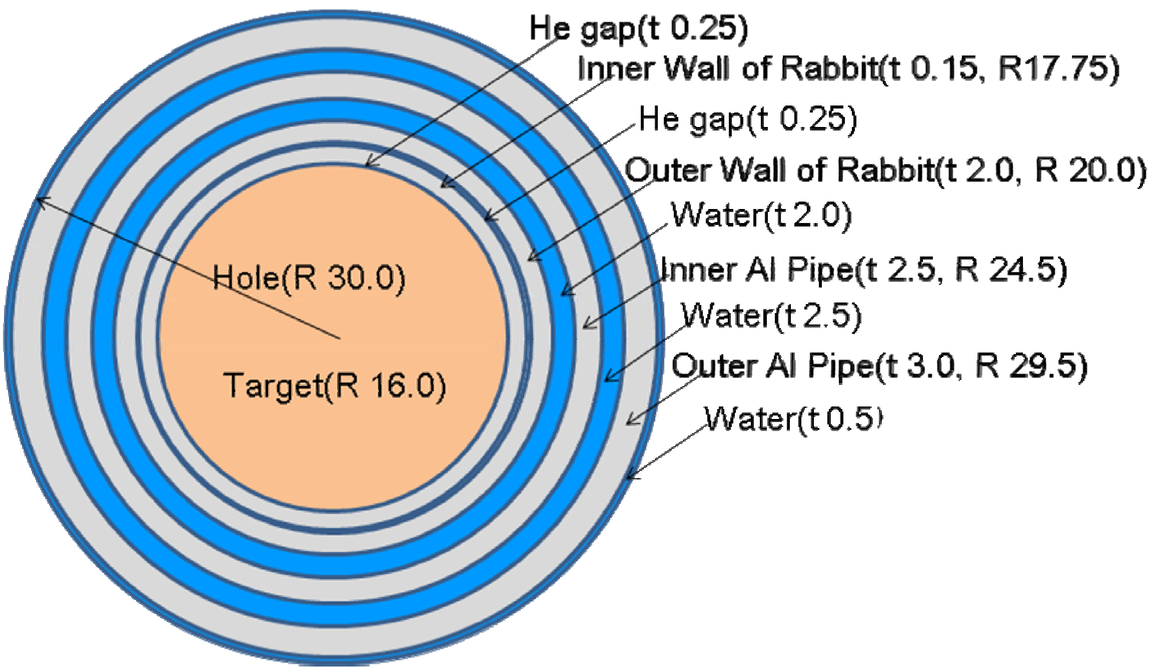

Figure 10. Horizontal dimensions for rabbit irradiation at OR3 (unit: mm)

APPENDIX: Regional demand for ^{99}Mo

A.1. Definition of the region

The region in this report includes areas within a few hours for the transportation of ^{99}Mo by frequent air lines, assuming that a regional cross backup supply scheme of ^{99}Mo is established. Potential countries to be benefited are Korea, Japan, China and Taiwan, which are sharing majority of ^{99}Mo demand in the Asia. Some other countries nearby such as Philippines, Vietnam, Mongolia and North Korea could also be benefited, but inclusion of ^{99}Mo demands in those countries is not necessary for this study because their demand is sufficiently small in the total. Among the four countries, Taiwan does not operate a reactor capable for meaningful production of ^{99}Mo, but they can be benefited from the scheme of regional ^{99}Mo supply if they are willing to.

A.2. Assessment of ^{99}Mo demand in the Asian region

Amount of ^{99}Mo for demand, supply or use is always expressed by its activity. As the ^{99}Mo decays with 66 h half-life, and its supply and use are long enough to change its activity significantly, the amount can be very different depending on a reference timing determining its activity. However, the amount is often expressed simply by its activity without any information on its reference timing. Data on ^{99}Mo demand of Korea and Japan are typical cases. Therefore, an assessment on the ^{99}Mo demand is needed to obtain meaningful data on the regional demand.

The status ^{99}Mo supply in the East and South-East Asia region surveyed through Forum for Nuclear Cooperation in Asia (FNCA) [A-1] is reproduced in Table A-1 with additional information for domestic production methods. Here, the demand is expressed by ^{99m}Tc not by ^{99}Mo. Total annual demand of the region is about 135 kCi/y when about 5 kCi/y is assumed for the demand of Taiwan, which corresponds to about 2,600 Ci/w. Japan and China share about 60 and 30%, respectively. However, it is not clear how the demand of each country in the table is defined. First of all, the demand in ^{99m}Tc has a different value from that in ^{99}Mo. Since the half-life of ^{99}Mo is about 11 times of ^{99m}Tc and branching ratio of ^{99m}Tc is about 88%, total activity of ^{99m}Tc produced from ^{99}Mo of initial activity 1 Ci is about 9.7 Ci. Actual activity of ^{99m}Tc utilized by a hospital is very dependent on the efficiency in the use.

For the demand of ^{99}Mo, the unit six-day-Ci is popularly used. The six-day-Ci is defined from a view point of ^{99}Mo solution suppliers, i.e., activity calibrated six days after shipment of ^{99}Mo by a supplier. It seems that an average time interval from the shipment to the use of ^{99}Mo is about six days for short distance supply. A supplier usually determines the shipping date twice a week at 12:00 local time of supplier facility. One shipment is for the use of ^{99}Mo from Monday and another one for the use from Wednesday or Thursday. The ^{99}Mo is finally used by ^{99m}Tc generators for a week; from Monday to Friday, from Thursday to next Wednesday, etc. It may also be used for two weeks, but the activity during the second week is only 17 % of the first week. A certain lead time is needed before a ^{99m}Tc generator is ready for use. For example, the ^{99}Mo solution is shipped on Thursday, ^{99m}Tc generators are made on Friday and generators are delivered for their use from next Monday. In this case, if there is no time difference between supplier and user, ^{99}Mo activity by the six-day-Ci matches with its activity at 12:00 Wednesday – middle of Monday and Friday. Therefore, the activity by the six-day-Ci is close to an average activity available to the user.

For the case of using 1 Ci ^{99m}Tc generator calibrated at 12:00 Wednesday, if the ^{99m}Tc is obtained at 09:00 every working day with 90% elution efficiency and then used for administration at 11:00, the total ^{99m}Tc available for administration for one working week (from Monday to Friday) is about 4 Ci. In this case, the demand of ^{99}Mo calibrated at 12:00 Wednesday (six-day-Ci) is about 1/4 of ^{99m}Tc demand. If the efficiency is increased by eluting twice in a day, it can be less than 1/4. In actual practice, however, since there are several other factors decreasing the efficiency, actual ^{99}Mo needed would be more than 1/4 of the ^{99m}Tc demand.

As cases of Korea and Japan, if the delivery of ^{99}Mo to users takes long time and time difference from the ^{99}Mo solution suppliers is also long, actual activity available for users becomes less. Other data on the demand of Korea and Japan are analyzed in the next section to understand actual demand in comparison with Table A-1.

A.3. Target for regional ^{99}Mo supply

The regional demand is a reference for the decision of production capability to be achieved for a regional supply of ^{99}Mo. Then a demand for the actual use has to be understood. In order to prevent confusion on the amount for the demand, it is assumed that the ^{99}Mo is available from the beginning of working week (09:00 Monday) and its activity is calibrated at middle of the working week (12:00 Wednesday). The ^{99}Mo can be supplied for its use from Wednesday or Thursday as well. In such case, for a same activity on the first day, available ^{99}Mo to a user becomes less due to its decay during weekend. However, such difference is neglected.

A radiopharmacy in Japan operating Master milkers informed that a 500 Ci Master milker calibrated at 12:00 Friday shares about 20% of domestic demand. It means that a total demand of Japan calibrated at 12:00 Friday is about 2,500 Ci/w. In June 2008, Koichi Iimura et al [A-2] estimated an annual demand of ^{99}Mo in Japan about 1,110 TBq/y (22.2 TBq/w = 600 Ci/w). They also mentioned that the demand is fulfilled by importing about 4,440 TBq/y (88.8 TBq/w = 2,400 Ci/w) from abroad. Similar expression is also found at the Summary Report of the FNCA 2006 Workshop on the Utilization of Research Reactor [A-1] – "*Net consumption of*

^{99}Mo in Japan is 600 Ci/w, however, decay compensated actual importing amount is 4 times, i.e. 2,400 Ci/w."

If it is assumed that all above data are approximately correct but different due to different timing in activity calibration, they may be understood as following: The amount of import (2,400 Ci/w) matches with the 2,500 Ci/w calibrated at 12:00 Friday. At 12:00 next Wednesday, it becomes 680 Ci, which approximately matches with above 600 Ci/w. Therefore, we can conservatively suppose that the actual demand in Japan is about 700 Ci/w when it is calibrated at 12:00 Wednesday.

For the case of Korea, the demand of ^{99m}Tc in the table is 300 Ci/w, but 200 Ci/w ^{99}Mo has been usually assumed. However, a recent announcement of Korean Government (see Section 2) was 110 Ci/w ^{99}Mo. Figure A-1 shows statistics on annual imports obtained from annual reports of Korea Radioisotope Society. Since there have been changes in the ^{99}Mo supply scheme in Korea, the graph cannot represent a consistent variation of ^{99}Mo import. Actual demand would have been almost steadily increasing until 2008, and 10,000 Ci/y (about 200 Ci/w) was expected soon. The import in 2009 of which data point was available very recently reduced to about 2/3 of 2008, which corresponds to about 120 Ci/w and approximately matches with the 110 Ci/w announced by the Government. Among the 110 Ci, 90 Ci is imported from SAFARI. Actually some more than 400 Ci is shipped on 12:00 Wednesday at SAFARI site (19:00 Wednesday in Korea), which is equivalent to 90 six-day-Ci. The shipping date and time is fixed by the policy of supplier.

From above information, it is confirmed that the unit Ci used in statistics for annual ^{99}Mo demand in Korea has been six-day-Ci but their shipping timing can be different depending on suppliers. Above announcement of Government also indicated that, even the import in 2009 was reduced to about 2/3 of previous year, there is no significant shortage in the use of ^{99m}Tc in Korea. How the demand can be satisfied by much reduced import may be explained that the efficiency in the use of ^{99}Mo is improved. Actually, if previous practices were not optimized ones, the efficiency can be increased by several ways such as changing shipping date of ^{99}Mo solution, multiple elutions of a ^{99m}Tc generator in a day, reducing time interval between elution and administration, etc. Anyway, current demand of Korea can be conservatively estimated as about 100 Ci/w when it is calibrated at 12:00 Wednesday. This also indicates that the ^{99}Mo imported to Korea had been decayed further to about a half compared to countries near from ^{99}Mo solution suppliers. It would be similar case for the Japan.

There is no other information for the demand of China. From number of nuclear medicine centers in the table and newly estimated demands of Korea and Japan above mentioned, the demand of China calibrated at 12:00 Wednesday is estimated about 550 Ci/w. Then total weekly demand of above three countries becomes 700 (Japan) + 100 (Korea) + 550 (China) = 1,350 Ci/w. Therefore, 1,500 Ci/w calibrated at 12:00 Wednesday can be safely set as a target of supply for the region including Taiwan and other nearby countries.

References

[A-1] Development of PZC-based Tc-99m Generator, Issue-2, Forum for Nuclear Cooperation in Asia (FNCA), March 2007, Edited by E.Z. Sombrito and T. Genka.

[A-2] Koichi Iimura, et al, Conceptual study of ^{99}Mo production facility in JMTR, JAEA-Technology, 2008-035, June 2008, in Japanese.

Table A-1. Status of ^{99}Mo demand and supply in East and South-East Asia region [A-1]

	Status of ^{99}Mo supply	Domestic production method	^{99m}Tc Demand (kCi/y)	No. of gamma cameras	No. of nuclear medicine centers
Japan	Import	[n,γ / solid]* [n,γ / liquid]*	75	1946	1257
China	Import/ Domestic	n,f + n,γ / solid	32	750	860
Korea	Import/ Domestic	n,γ / solid	15	280	140
Malaysia	Import	-	1	18	14
Philippines	Import	-	1	23	25
Vietnam	Import/ Domestic	n,γ / solid	0.7	16	24
Thailand	Import	-	0.5	32	21
Indonesia	Domestic +Export	n,f	0.4	32	16
Bangladesh	Import	-	0.2	22	17

* under plan

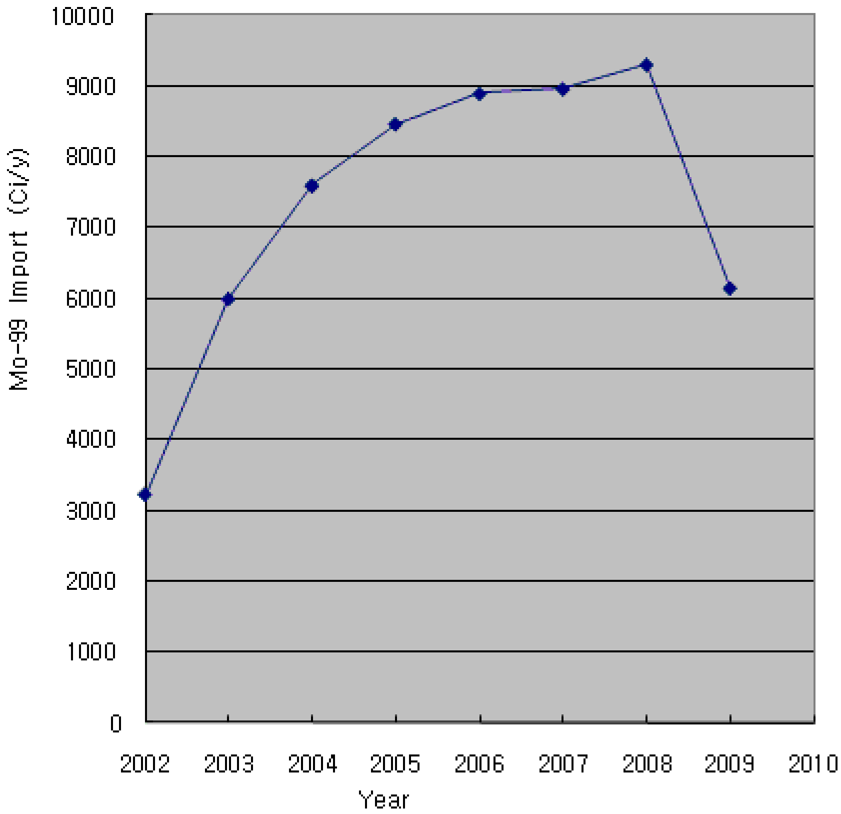

Figure A-1. Trend of annual ^{99}Mo import of Korea

* Much less imports up to 2004 may be due to inconsistency in data

* Sudden drop in 2009 is due to shortage of supply

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^2
体積	立法メートル	m^3
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^2
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^3
面積密度	キログラム毎平方メートル	kg/m^2
比体積	立方メートル毎キログラム	m^3/kg
電流密度	アンペア毎平方メートル	A/m^2
磁界の強さ	アンペア毎メートル	A/m
量濃度 (a)，濃度	モル毎立方メートル	mol/m^3
質量濃度	キログラム毎立法メートル	kg/m^3
輝度	カンデラ毎平方メートル	cd/m^2
屈折率 (b)	（数字の） 1	1
比透磁率 (b)	（数字の） 1	1

(a) 量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。
(b) これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン (b)	rad	1 (b)	m/m
立体角	ステラジアン (b)	sr (c)	1 (b)	m^{2}/m^2
周波数	ヘルツ (d)	Hz		s^{-1}
力	ニュートン	N		$m\ kg\ s^{-2}$
圧力，応力	パスカル	Pa	N/m^2	$m^{-1}\ kg\ s^{-2}$
エネルギー，仕事，熱量	ジュール	J	N m	$m^2\ kg\ s^{-2}$
仕事率，工率，放射束	ワット	W	J/s	$m^2\ kg\ s^{-3}$
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	$m^2\ kg\ s^{-3}\ A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2}\ kg^{-1}\ s^4\ A^2$
電気抵抗	オーム	Ω	V/A	$m^2\ kg\ s^{-3}\ A^{-2}$
コンダクタンス	ジーメンス	S	A/V	$m^{-2}\ kg^{-1}\ s^3\ A^2$
磁束	ウエーバ	Wb	Vs	$m^2\ kg\ s^{-2}\ A^{-1}$
磁束密度	テスラ	T	Wb/m^2	$kg\ s^{-2}\ A^{-1}$
インダクタンス	ヘンリー	H	Wb/A	$m^2\ kg\ s^{-2}\ A^{-2}$
セルシウス温度	セルシウス度 (e)	℃		K
光束	ルーメン	lm	cd sr (c)	cd
照度	ルクス	lx	lm/m^2	$m^{-2}\ cd$
放射性核種の放射能 (f)	ベクレル (d)	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	$m^2\ s^{-2}$
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト (g)	Sv	J/kg	$m^2\ s^{-2}$
酸素活性	カタール	kat		$s^{-1}\ mol$

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。
(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。
(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。
(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。
(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。
(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で"radioactivity"と記される。
(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	$m^{-1}\ kg\ s^{-1}$
力のモーメント	ニュートンメートル	N m	$m^2\ kg\ s^{-2}$
表面張力	ニュートン毎メートル	N/m	$kg\ s^{-2}$
角速度	ラジアン毎秒	rad/s	$m\ m^{-1}\ s^{-1}=s^{-1}$
角加速度	ラジアン毎秒毎秒	rad/s^2	$m\ m^{-1}\ s^{-2}=s^{-2}$
熱流密度，放射照度	ワット毎平方メートル	W/m^2	$kg\ s^{-3}$
熱容量，エントロピー	ジュール毎ケルビン	J/K	$m^2\ kg\ s^{-2}\ K^{-1}$
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	$m^2\ s^{-2}\ K^{-1}$
比エネルギー	ジュール毎キログラム	J/kg	$m^2\ s^{-2}$
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	$m\ kg\ s^{-3}\ K^{-1}$
体積エネルギー	ジュール毎立方メートル	J/m^3	$m^{-1}\ kg\ s^{-2}$
電界の強さ	ボルト毎メートル	V/m	$m\ kg\ s^{-3}\ A^{-1}$
電荷密度	クーロン毎立方メートル	C/m^3	$m^{-3}\ sA$
表面電荷	クーロン毎平方メートル	C/m^2	$m^{-2}\ sA$
電束密度，電気変位	クーロン毎平方メートル	C/m^2	$m^{-2}\ sA$
誘電率	ファラド毎メートル	F/m	$m^{-3}\ kg^{-1}\ s^4\ A^2$
透磁率	ヘンリー毎メートル	H/m	$m\ kg\ s^{-2}\ A^{-2}$
モルエネルギー	ジュール毎モル	J/mol	$m^2\ kg\ s^{-2}\ mol^{-1}$
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	$m^2\ kg\ s^{-2}\ K^{-1}\ mol^{-1}$
照射線量（Ｘ線及びγ線）	クーロン毎キログラム	C/kg	$kg^{-1}\ sA$
吸収線量率	グレイ毎秒	Gy/s	$m^2\ s^{-3}$
放射強度	ワット毎ステラジアン	W/sr	$m^4\ m^{-2}\ kg\ s^{-3}=m^2\ kg\ s^{-3}$
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2\ sr)$	$m^2\ m^{-2}\ kg\ s^{-3}=kg\ s^{-3}$
酵素活性濃度	カタール毎立方メートル	kat/m^3	$m^{-3}\ s^{-1}\ mol$

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10800) rad
秒	”	1”=(1/60)’=(π/648000) rad
ヘクタール	ha	$1ha=1hm^2=10^4m^2$
リットル	L，l	$1L=1l=1dm^3=10^3cm^3=10^{-3}m^3$
トン	t	$1t=10^3$ kg

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	1eV=1.602 176 53(14)×10^{-19}J
ダルトン	Da	1Da=1.660 538 86(28)×10^{-27}kg
統一原子質量単位	u	1u=1 Da
天文単位	ua	1ua=1.495 978 706 91(6)×10^{11}m

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	1 bar=0.1MPa=100kPa=10^5Pa
水銀柱ミリメートル	mmHg	1mmHg=133.322Pa
オングストローム	Å	1 Å=0.1nm=100pm=10^{-10}m
海里	M	1 M=1852m
バーン	b	1 b=100fm^2=(10^{-12}cm)2=$10^{-28}m^2$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デジベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	1 erg=10^{-7} J
ダイン	dyn	1 dyn=10^{-5}N
ポアズ	P	1 P=1 dyn s cm^{-2}=0.1Pa s
ストークス	St	1 St =1$cm^2\ s^{-1}$=$10^{-4}m^2\ s^{-1}$
スチルブ	sb	1 sb =1cd cm^{-2}=10^4cd m^{-2}
フォト	ph	1 ph=1cd sr cm^{-2} 10^4lx
ガル	Gal	1 Gal =1cm s^{-2}=$10^{-2}ms^{-2}$
マクスウェル	Mx	1 Mx = 1G cm^2=10^{-8}Wb
ガウス	G	1 G =1Mx cm^{-2} =10^{-4}T
エルステッド (c)	Oe	1 Oe ≙ (10^3/4π)A m^{-1}

(c) ３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	1 Ci=3.7×10^{10}Bq
レントゲン	R	1 R = 2.58×10^{-4}C/kg
ラド	rad	1 rad=1cGy=10^{-2}Gy
レム	rem	1 rem=1 cSv=10^{-2}Sv
ガンマ	γ	1 γ =1 nT=10-9T
フェルミ		1フェルミ=1 fm=10-15m
メートル系カラット		1メートル系カラット = 200 mg = 2×10-4kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー）4.184J（「熱化学」カロリー）
ミクロン	µ	1 µ =1µm=10^{-6}m

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