RECENT ACTIVITIES OF KAERI RELATED TO FISSION MO-99

C.K. KIM, I.C. Lim, K.H. KIM, H.J. RYU, J.M. PARK, S.J. CHOI

Research Reactor Fuel Development, KAERI
Daedeokdaero 1045, Yuseong, Daejeon, 305-353, Republic of Korea

ABSTRACT

Relating to the fission Mo in KAERI there are three activities which are a new research reactor for producing Mo-99, a development of uranium foil for using annular type fission Mo target, and a new target development using uranium metal powder produced by atomization technology, which was developed by KAERI. The project of new research reactor was approved thorough a feasibility study conducted by Korea Development Institute. The design and construction will be carried out from 2012 to 2016. The major utilization targets are radio-isotope production and neutron transmutation doping. The majority of isotope productions will be produced in this reactor including Mo-99, and the capacity will be enough to fulfil the national demand and for exportation to regional countries. The newly proposed target of atomized uranium particles dispersion plate type was estimated to be 8.5 g-U/cm^3 supposing the volume fraction of the dispersion to be 45 Vol.%. When the HEU fission Mo target of 1.5 g-U/cm^3 is converted to LEU, a LEU uranium density of more than 7.5 g-U/cm^3 is required. The atomized U metal particles dispersion plate target of more than 8.5 g-U/cm^3 would be applicable. It is considered that the target would maintain the integrity from aspects of very short time irradiation and very fine grain. In order to improve the foil quality and productivity an equipment was designed newly under the various concepts and manufactured successfully with domestic industry assistance in last year. In this year efforts were made to complement the weak points appeared in the preliminary experiments. A sound uranium foil could be produced by adapting cooling jacket pad, forcing down the forming foil by Ar gas injection, accurately controlling the clearance between melt feeding nozzle tip and casting roll surface.

1. Introduction

KAERI launched a project of constructing of multi-purpose research reactor, which is now HANARO, around the middle of 1980s. The reactor was designed the fuel to be LEU U_3Si-Al dispersion rod type fuel. Several years later a project of developing the fuel was established for localizing. The construction of HANARO was finished around the middle of 1990s and the start-up operation was done in 1995. Since that time HANARO is devoting for neutron scattering experiments, irradiation tests of material and fuel for power reactor, RI production, silicon doping, neutron activation analysis, neutron radiography. The fuel development was successfully carried out with implementing a new process of atomization technology in making fuel powder uniquely. Some fuel bundles were fabricated for the irradiation tests in conjunction with fuel qualification around the latter part of 1990s. Two times of irradiation tests for mini-elements and full demonstration of irradiation for full size bundle were conducted and the fuel fabrication facility was designed and constructed around 2004. Nowadays the fuels are fully domestically provided for HANARO.

The technologies related to design, operation, and utilization have been accumulated through above experiences in the mean time and then enabled to participate in some international biddings for constructions of new research reactors such as the Pallas project and the JRTR project. Fortunately KAERI was awarded the bidding of the JRTR project. The design for the JRTR is underway. The JRTR is planning to use U-Mo dispersion plate type fuel. KAERI has U-Mo powder production technology by atomization method but no experience in the plate type fuel fabrication. This situation raised a plan of constructing a U-Mo dispersion plate fuel production line using atomization technology for U-Mo power. In Korea radio-isotope of Mo-99 has been imported and experienced a Mo-99 shortage. Also the need for silicon transmutation doping service is increasing. These things have motivated KAERI to consider a new research reactor.

Currently HEU minimization efforts in fission Mo production are underway in connection with the global threat reduction policy. In order to convert HEU to LEU for the fission Mo target, higher uranium density material is needed [1]. From the stable behaviour during irradiation, UAlx and UO_2 have mainly been used as a fission Mo target material [2]. The HEU UO_2 annular target could easily be replaced with the uranium foil target developed by ANL. This study focused on the dispersion plate target, which is very similar to the dispersion plate fuel of research reactor. The uranium aluminide targets used world widely for commercial ^{99}Mo production are limited to 3.0 g-U/cc in uranium density of the target meat [3]. A high uranium density using the uranium metal particles dispersion plate target is raised with taking an advantage of the atomization technology developed by KAERI for powdering U or U alloy [4]. The irradiation burnup of the target is very low as 6 % of fission fragments. The irradiation period is shorter than 7 days. It is considered that the degradation by irradiation would be allowable.

In conjunction with uranium foil for higher uranium density KAERI developed an alternative process of directly forming foil from the melt by roll casting for LEU foil for the purpose of simplifying the fabrication process as well as saving the cost around the beginning of 2010s. Some foils were distributed to various countries, which are Chile, Indonesia, Romania, Poland, and USA, under CRP of the IAEA. Especially for Libya some assembled targets was provided. A drawback on KAERI foil was issued. The foil thickness is so inhomogeneous that some difficulties would be possibly suffered in applying to fission Mo target. KAERI established a new project on improving the foil quality as well as the fabrication efficiency in 2008. The main concept was an adaption of self-gravity flowing casting system using graphite crucible system with avoiding the use of fragile quartz crucible, which is very sensitive in holding the melt under pressure. A new equipment was designed under the above concept and manufactured successfully with domestic industry assistance. Some efforts on improving the foil quality, yield, and the productivity were made through experiments.

In this paper three kinds of activities relating to the fission Mo are described.

2. New Korea Research Reactor

The designed major characteristics for the new research reactor are as follows:

- Reactor power: ~20 MW
- Reactor type: Pool type
- Maximum thermal flux: > 3.0×10^{14} n/cm^{2}s
- Operation: ~300 days/year
- Fuel: LEU U-Mo Plate type fuel
- CEDM (Control Element Driving Mechanism) location: bottom of core
- Reflector: Beryllium and Graphite
- Reactor life: 50 years

A feasibility study was conducted by KDI (Korea Development Institute) and appeared to be positive. The related fund will be provided through an approving process of the nation assembly from next year.

The site is a place in Gijang-gun, which is a county near Busan city. There are several nuclear power reactors nearby. The proposed site was chosen through an evaluation of proposals from many counties to host the facility. The construction project is expected to start from Jan 2012, and will finish by the end of 2016.

The major utilization targets are radio-isotope production and neutron transmutation doping. The majority of isotope productions will be produced in this reactor including Mo-99, and the capacity will be enough to fulfil the national demand and for exportation to regional countries. It is not easy to foresee the future utilization needs during the reactor life time. Considering this, the reactor will be designed to accommodate the mid-term irradiation service needs, and to be flexible for future changes in use.

When the design is realized, the reactor will be the first research reactor to use U-Mo fuel for real operation. After the completion of the project, the irradiation service functions of HANARO will be turned over to the new reactor, and HANARO will focus on neutron scattering experiments and fuel/material development.

3. Consideration of A Very High Uranium Density Plate Target

In KAERI, the atomization technology for producing uranium silicide as well as U-Mo powders had been developed in 1990s. In the mean time some pure uranium metal powder has been produced many times to provide for advanced nuclear fuel development. So it is considered that the pure uranium metal powder can be supplied without any difficulties. The most fission Mo targets of fission Mo commercial producers were reported to be UAlx dispersion plate type. In the fabrication process the HEU UAlx powder could be replaced with atomized LEU metal powder without any difficulties. It is considered that they need a small modification.

In the aspect of the fabrication process, the existing fabricators can adapt this atomized particles dispersion process if atomized U powder is supplied effectively [5]. The additional steps such as the mixing of u powder and Al powder and compacting are very common and proven technology in research reactor fuel fabrication. Presumably the cost could not increase much for adapting the additional steps.

The average volume of U_3Si_2 dispersion fuel by rolling work was about 45 Vol.%. If the volume fraction is supposed to be 45 Vol.% in case of atomized spherical particles powder, the available uranium density would be 8.5 g-U/cm^3. When the HEU fission Mo target of 1.5 g-U/cm^3 is converted to LEU, a LEU uranium density of more than 7.5 g-U/cm^3 is required. It is considered that the atomized U metal particles dispersion plate target of more than 8.5 g-U/cm^3 would be applicable.

In general the spherical particles powder with spherical shape has better plasticity in rolling work than irregular particles powder due to a smooth surface. The average fabrication porosity of research reactor fuel dispersed with atomized particles appeared to be as small as less than 3 Vol.% from the better plasticity [5].

The fission Mo target should maintain its integrity during the irradiation. The target center temperature was calculated by using the PLATE computer code developed by ANL by supposing that the heat flux and cooling water speed are 250 W/cm^2.and 6 m/sec, respectively. The thermal conductivity of pure uranium metal is fairly good. So that average thermal conductivity of atomized particles dispersion was estimated to be 85 W/m-K. The temperature increases for the uranium particles dispersion target with a thickness of 1.0 mm and cladding with a thickness of 0.3 mm were calculated to be 4.5 °C and 1.5 °C, respectively. The temperature increase at the interface between the cladding and the coolant water was about 25 °C. When the coolant temperature at the outlet is supposed to be 40 °C, the peak temperature at the target center is estimated to be 71 °C. It is considered that these values have enough safe margins in the aspect of the thermal induced swelling. The center temperature of about 71 °C is presumed to be too low to induce an interaction at the interface between the uranium particles and the aluminium matrix.

The atomization process has an advantage of a very rapidly solidification in forming particles. So the grains inside the particles are very fine. The grain sizes for atomized U particles were measured to be a few microns while the grain sizes of the uranium common cast ingot were measured several hundred microns. It has been known that some alloying elements of Al, Si, Cr, Fe, and Mo are beneficial for reducing the swelling by grain refinement. In uranium metal fuel the irradiation behaviour could be improved by alloying some elements of Fe, Si, Al, Cr, and Mo [6]. The dispersed uranium particles containing those kinds of alloying elements can be easily made by adding the alloying elements into the crucible when the melting operation is prepared. The fission Mo target is irradiated less than 8 at% burnup. As shown above, the fission target temperature is much lower than the metal fuel temperature due to very thin thickness. The irradiation period of the fission target in the reactor is much shorter than that of uranium metal fuel. It is considered that the integrity of the atomized uranium particles dispersion target containing some grain refining elements could be maintained up to the aiming irradiation.

In developing U-Mo dispersion fuel for high performance research reactors an interaction problem between dispersed U-Mo particles and Al matrix occurs. A silicon addition to an aluminium matrix was found to give a good effect on retarding the interaction rate. If it is supposed that the interaction at the interface between uranium particles and the aluminium matrix occurs, the dispersion target temperature would increase. Then the thermal conductivity would decrease gradually. As the irradiation continues, the temperature increasing rate would be accelerated. However, if the calculated

temperature is rightly estimated, such a low temperature would not approach the accelerating stage criteria.
In dissolving the steps for separating 99Mo from the solution, the fission target can be put into the dissolving vessel. Because the target is not opened, any leakage radioactive gas would not take place. If the aluminium matrix and cladding is dissolved by NaOH and the solution containing aluminium is decanted, aluminium material can be separated as a low level- waste. Then the remaining particles can be dissolved by HNO_3. The following steps are the same as the CITICHEM process. Most of fission products are contained in the uranium particles. The high radioactive waste would be produced as a small quantity.

4. Progress on Development on U Foil Fabrication Technology in KAERI

The new designed and manufactured equipment was installed in KAERI around July 2010 and then some preliminary results were reported in the last RERTR meeting. The status related to this matter was summarized as following.

1) The foil surface appeared to exist some irregularity on the upper free surface
2) The foil thickness was measured to be thicker than 200 μm
3) The foil produced by new equipment was more brittle comparing to foils by the previous quartz crucible equipment.
4) The yield of sound foil was lower than that by the previous quartz crucible equipment.

In order to overcome the above weak points some complementary measures were conducted as following [7].

1) Improving the thickness homogeneity
   - Some modifications were done for melt to be fed more steadily with less eddy flow.
2) To reduce very thick layer of more than 200 μm
   - Higher revolution speed for casting roll was applied. In case of too high revolution speed, foil is liable to outward flying by centrifugal force. The outward flying action is suppressed by high pressure argon gas injecting to the formed foil attached to the roll surface.
   - Minimizing the clearance of slit nozzle tip and roll surface
3) To overcome the brittle property of foil
   - Large volume chamber of new equipment is difficult in controlling vacuum-tight controlling
   - Additional installation of diffusion vacuum pump
   - Precisely vacuum control with accurate maintenance for connections of components and pipes.
4) To pursuit higher yield of sound thin foil
   - Phenomenon of splitting and breaking to pieces in foil formation
   - Enforcement of solidifying efficiency by installing cooling pad at revolving casting roll to cool the periphery part of casting roll heated by very closely located heater for tundish
   - Installation of argon gas injection onto the forming foil at roll to preventing outward flying and the winding foil for more efficient collection

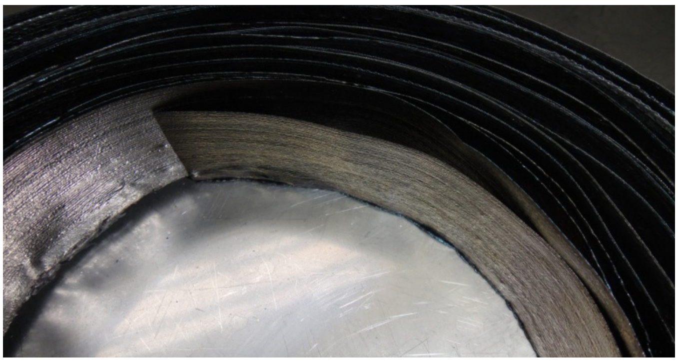
Recently a fairly good foil was produced as shown in Figure 1. Total length and its thickness are about 10 meters and 135 -145 μm, respectively as in Table 2. This thickness is very close to the requiring thickness of 135 μm of the Cintichem process developed by ANL. When higher revolution speed In order to get thinner foil was applied, the revolution speed was limited to about 60 rpm due to splitting occurrence in foil and outward flying phenomenon. The effective parameter for controlling the thickness is considered to be not only revolution speed of casting roll but also clearance between melt feeding nozzle tip and casting roll surface.
The thickness homogeneity of foil did not get improved much. However, the upper free surface of solidifying foil was changed from short range regularity to longer range regularity as shown in Figure 3. The cooling jacket pad is considered to give a positive effect on forming sound foil without any holes by inducing more quickly solidifying the fed melt layer.
When the vacuum degree got improved by additional installation of diffusion vacuum pump and the

precise maintenance for connection parts, the foil appeared more ductile with having luster surface. The argon gas injection on to the coming out foil appeared to be effect on forcing down the forming foil

Fig. 1 Recently produced Foil

Table 1. Thickness measurements for recently produced foil

	Front end part	Middle part (5m from front tip)	Tail part (10 m from front tip)
width(mm)	6.5	5.2	4.2
Thickness	199	182	167
	198	160	136
	221	242	198
	172	176	203
	119	169	124
	150	120	107
	158	133	120
	136	146	136
	131	131	127
	139	142	146
average(μm)	135	139	145
Standard Deviation	34	35	33



Previous

Present

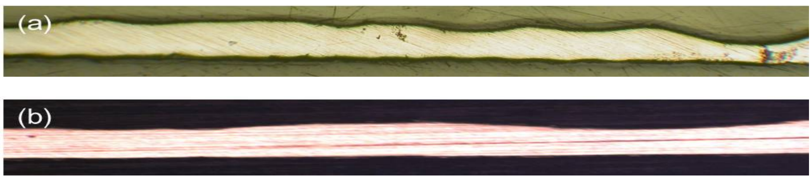


Fig. 3 Cross Section of Uranium Foil

to the casting roll surface and made the foil wound properly. The tip speed of winding machine and the tip speed of casting roll were difficult to match. The foil appeared broken with some disconnections.
In near future a rolling machine is scheduled to install in connection with the plate type fuel fabrication facility construction in KAERI. Presumably a rolling work to straighten some irregular foil would be possible. Some more precisely thickness-controlled U foil could be available.

6. Summary

A feasibility study for the new Korean Research Reactor was conducted by KDI (Korea Development Institute) and appeared to be positive. The related fund will be provided through an approving process of the nation assembly from next year. The construction project is expected to start from Jan 2012, and will finish by the end of 2016. The major utilization targets are radio-isotope production and neutron transmutation doping. The majority of isotope productions will be produced in this reactor including Mo-99, and the capacity will be enough to fulfil the national demand and for exportation to regional countries.
A sound uranium foil could be produced by adapting cooling jacket pad, forcing down the forming foil by Ar gas injection, accurately controlling the clearance between melt feeding nozzle tip and casting roll surface. The thin sound foil approaching to 135 μm without any holes was obtained. The foil yield was advanced with producing long foil of more than 10 meter. The foil roughness was not much improved. It is considered that a roll-work for the some irregular foil would be possible by the installation-scheduled roll machine in KAERI. Better quality foil could be available for fission Mo target presumably.

Reference

1) Parrish Staples, “Global Threat reduction Initiatives”, IAEA Consultancy Meeting, 2010
2) Chris Whipple et al. ”Medical Isotope Production without Highly enriched Uranium”, The national academies
3) Gavin Ball, ”Experiences of HEU to LEU Mo-99 Production Conversion at NTP”, IAEA Consultancy Meeting, 2010
4) C.K. Kim, et al. “A consideration for uranium high density plate type target of uranium metal particles dispersion in aluminium matrix for 99Mo production”, RRFM 2011, Rome.
5) C.K. Kim, J.M. park, H.J. Ryu, “Use of a Centrifugal Atomization Process in the Development of Research Reactor Fuel” Nuclear Engineering and Technology, Vol. 39, No. 5,
6) William R. McDonell, George R. Caskey, and Carl L. Angerman, “High-performance Uranium-Metal Fuels foe Savannah River Reactors”, WSRC-MS-2000-00061
7) C.K. Kim, et al. “Progress on Development on U Foil Fabrication Technology in KAERI”, RERTR 2011, Santiago.