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Conversion of Molybdenum-99 production process to low enriched uranium: Neutronic and thermal hydraulic analyses of HEU and LEU target plates for irradiation in Pakistan Research Reactor-1

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ABSTRACT

Technetium-99m, the daughter product of Molybdenum-99 is the most widely needed radionuclide for diagnostic studies in Pakistan. Molybdenum-99 Production Facility has been established at PINSTECH. Highly enriched uranium (93% ^{235}U) U/Al alloy targets have been irradiated in Pakistan Research Reactor-1 (PARR-1) for the generation of fission Mo-99, while basic dissolution technique is used for separation of Mo-99 from target matrix activity. In line with the international objective of minimizing and eventually eliminating the use of HEU in civil commerce, national and international efforts have been underway to shift the production of medical isotopes from HEU to LEU (LEU; <20% ^{235}U enrichment) targets. To achieve the equivalent amount of ^{99}Mo with LEU targets, approximately 5 times uranium is needed. LEU aluminum uranium dispersion target has been developed, which may replace existing HEU aluminum/uranium alloy targets for production of ^{99}Mo using basic dissolution technique. Neutronic and thermal hydraulic calculations were performed for safe irradiation of targets in the core of PARR-1.

1. Introduction

Technetium-99m (^{99m}Tc; $T_{½}$ = 6 h)), the daughter product of Molybdenum-99 (^{99}Mo; $T_{½}$ = 66 h), is the most widely needed radionuclide in the world, used for well over 30 million medical diagnostic procedures annually and comprising over 80% of all diagnostic nuclear medicine procedures. Until now, approximately 95% of ^{99}Mo is produced in research and material test reactors by irradiation of Highly Enriched Uranium (HEU; >20% ^{235}U enrichment, generally 93%) targets that are subsequently dissolved to recover fission ^{99}Mo. Research and development work on production of ^{99m}Tc and ^{99}Mo by accelerator [1–2] and n, gamma reactions have also been carried out [3]. In line with the international objective of minimizing and eventually eliminating the use of HEU in civil commerce, national and international efforts have been underway to shift the production of medical isotopes from HEU to LEU (LEU; <20% ^{235}U enrichment) targets. A small amount of the current global ^{99}Mo production is derived from the irradiation of LEU targets. CNEA Argentina successfully converted its ^{99}Mo production to LEU and has been routinely producing since 2002 [4]. BATAN, Indonesia has converted ^{99}Mo production to LEU, based on foil targets and the LEU-Modified Cintichem process with assistance from Argonne National Laboratory, USA [5]. New LEU-based ^{99}Mo commercial-scale production facility has been in operation at OPAL, constructed by INVAP, SE. (Argentina) based on CNEA targets and processing methods [6].

The Reduced Enrichment for Research and Test Reactors (RERTR) Program develops technology necessary to enable the conversion of civilian facilities using high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The RERTR Program was initiated by the US. Department of Energy in 1978. During the Program's existence, over 40 research reactors have been converted from HEU to LEU fuels, and processes have been developed for producing radioisotopes with LEU targets. Under RERTR initiative Pakistan Research Reactor-1 (PARR-1), a swimming pool MTR type research reactor which attained full power of 5 MW in June, 1966, with 93% high enriched uranium (HEU) fuel was converted to <20% LEU fuel in October, 1991. The reactor power was also upgraded from 5 to 9 MW and then to 10 MW in 1998 [7].

In foreseeable future, PARR-2 will also be converted to use LEU [8]. The study is underway as a part of the IAEA coordinated research project (CRP) entitled "Conversion of the Miniature Source Reactors (MNSR) to Low Enriched Uranium (LEU)".

Under IAEA coordinated research project (CRP) entitled "Establish techniques for small scale indigenous molybdenum-99

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production using LEU fission or neutron activation'', efforts to convert HEU target to LEU target is continued. PINSTECH is producing fission ^{99}Mo by irradiating HEU plate targets in the core of Pakistan Research Reactor-1.

An agreement between PINSTECH/Pakistan Atomic Energy Commission and International Atomic Energy Agency Safeguards Department was signed in 2010. Safeguards Approach narrates the ''Applications of Safeguards to HEU/Al Target Plates for Mo-99 Production at PARR-1''. At the 'Molybdenum-99 Production Facility' the irradiated targets plates will be subjected to alkaline digestion and the uranium, plutonium produced and fission products will precipitate while Mo-99 and some fission products will remain in solution. The Mo-99 will be purified by chromatography and sublimation techniques and the remaining irradiated precipitate (solid phase) containing uranium-235 (4.63 g), and actinides with fission product will be put into a small stainless steel container which will be transferred to a segregated location in the 'Spent Fuel Storage Bay' of PARR-1 where it will be stored out of safeguards. Before the accumulated amount of material on which safeguards has been suspended reaches the permissible limit of 1000 g, safeguards will be reapplied to the HEU residue and the small containers verified before loading into a large stainless steel container. The large container will be removed from the bay, drained and decontaminated as necessary before being sealed by the Agency. Once the HEU residue is enclosed in the large container it will no longer be available for verification and will therefore be considered as difficult-to-access. Safeguards will be applied to fresh HEU/AI target plates if stored at PARR-1 prior to irradiation.

To achieve the equivalent amount of ^{99}Mo with LEU targets, approximately 5 times uranium is needed. LEU Aluminum uranium dispersion target has been developed, which may replace existing HEU aluminum/uranium alloy targets for production of ^{99}Mo using basic dissolution technique.

Different thermal hydraulic limiting phenomena's are important to be studied during the natural convection cooling of a hypothetical irradiated Low Enriched Uranium (LEU) target [9–11]. Neutronic and thermal hydraulic calculations were performed for safe irradiation of targets in the core of PARR-1. The results of HEU and LEU targets are compared.

2. Pakistan Research Reactor

2.1. History

The Pakistan Research Reactor-1 (PARR-1) is a swimming pool type research reactor originally designed for a thermal power of 5 MW in 1968. Its core has been redesigned to operate with LEU fuel at a power level of 9 MW in 1992 and 10 MW in 1998. PARR-1 is mainly utilized for radioisotope production, neutron activation analysis and material structure testing.

	1	2	3	4	5	6	7	8	9
A			GR-14	S-109	S-103	S-93	S-88	S-97	S-96
B		GR-16	GR-10	S-108	S-98	SR#5 C-26	S-70	SR#1 C-27	S-83
C		FC-A	GR-19	WB-7	S-92	S-67	WB-3	S-89	S-87
D		GR-07	WB-2	S-107	SR#4 C-25	S-86	S-85	S-68	SR#2 C-23
E			GR-03	S-106	S-101	S-90	SR#3 C-24	S-95	S-99
F		FC-B		S-105	S-104	S-91	S-94	S-100	S-102

Thermal Column

S-XX	Standard Fuel Element
C-XX	Control Fuel Element
FC-A, FC-B	Fission Chambers
WB-XX	Water Box
GR-XX	Graphite Reflector

Fig. 1. Core loading No. 100 for irradiation of LEU target plates.

Table 1
Specifications of HEU and LEU plate targets.

Characteristic	HEU plate	LEU plate
Outer length	160 mm	160 mm
Outer width	60 mm	60 mm
Outer thickness	1.3 mm	1.5 mm
Meat length	125 mm	125 mm
Meat width	40 mm	40 mm
Meat thickness	0.5 mm	0.7 mm
Clad thickness	0.4 mm	0.4 mm
Fuel material	U–Al	UAl_2–Al
Uranium content	1.667 g	7.595 g
^{235}U Enrichment	93%	19.75%
Al weight (maximum)	32 g	33 g

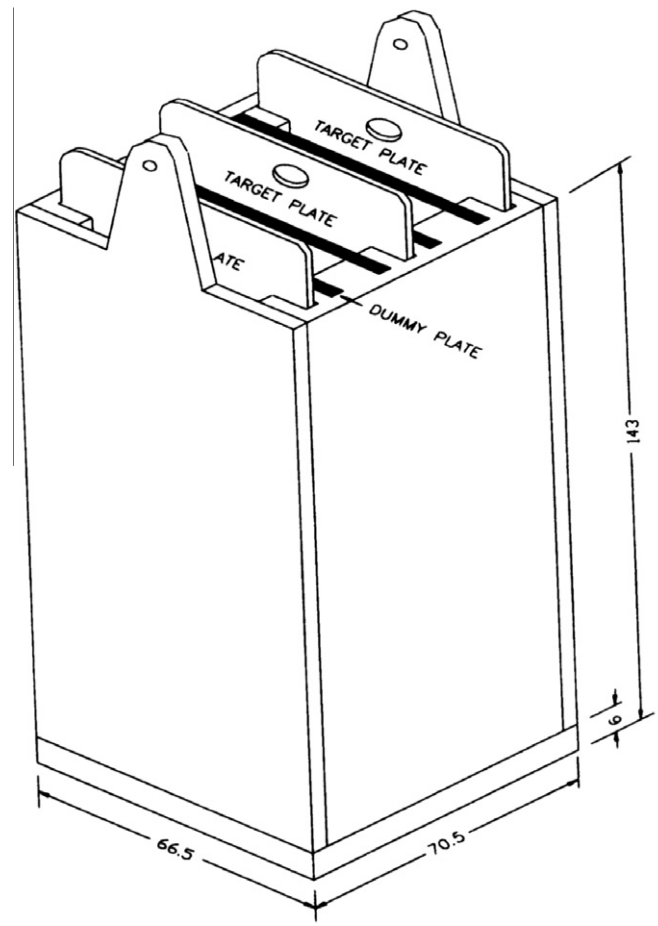

Fig. 2. Target fuel plate holder (All dimensions in mm).

2.2. PARR-1 core assembly

The PARR-1 core consists of an assembly of standard and control fuel elements mounted on the grid plate. The fuel elements can be assembled in different core configurations. The core is immersed in demineralized water which acts as coolant, moderator and reflector. However, using specially designed reflector elements the light water can be replaced on one or more sides with other reflectors such as heavy water, graphite, or beryllium.