EGYPT FINAL REPORT FOR THE CRP ON "DEVELOPING TECHNIQUES FOR SMALL- SCALE, INDIGENOUS PRODUCTION OF MO-99 USING LOW-ENRICHED URANIUM (LEU) OR NEUTRON ACTIVATION"

1. INTRODUCTION

The Egypt country report presented here describes the CRP activities at Egypt's second research reactor (ETRR-2) during the time period from 15-09-2007 to 30-11-2011. This chapter contains the existing capabilities and facilities and a description of the research carried out and achievements. Some of the results of calculations and start up tests are also presented.

2. EAEA EXISTING CAPABILITIES AND FACILITIES

The Egyptian Atomic Energy Authority (EAEA) has the following capabilities and facilities in support of production of ⁹⁹Mo using LEU plates or neutron activation:

2.1. ETRR-2 RESEARCH REACTOR

The ETRR-2 is an open pool Material Testing Reactor (MTR) and achieved first criticality on November 1997. It is using low enrichment plate fuel element (less than 20% enrichment), and is cooled and moderated by light water and reflected by beryllium and water. The reactor power is 22 MW with high neutron flux irradiation.

The reactor has two fast irradiation positions, two silicon irradiation positions, three radial and one tangential beam tubes, and thermal column. Several experimental and production facilities have been installed for radioisotope (RI) production (¹³¹I, ¹²⁵I, ⁵¹Cr, ¹⁹²Ir, and ⁶⁰Co), neutron activation analysis (NAA) applications, neutron transmutation doping (NTD), neutron radiography experiments, and training of personnel. A special hot cell for irradiated material testing has been installed where the impact test, tensile tests, and other material characterization can be applied for irradiated material samples.

2.2. RADIOISOTOPE PRODUCTION DEPARTMENT

This department is equipped with necessary hot cell, chemical processing and production of 99 Mo from neutron activation of 98 Mo, which is supplied in cooperation with Chinese experts.

2.3. NEW RADIOISOTOPE PRODUCTION FACILITIES (RPF)

The RPF is located in the same site of the ETRR-2 reactor. The RPF is a contract with INVAP SE (Argentina) to produce ⁹⁹Mo by irradiation of LEU at ETRR-2 reactor. The RPF was commissioned during October and November 2011. It is equipped with necessary hot cells and equipments for production of ⁹⁹Mo from the irradiated LEU and loading of ^{99m}Tc.

2.4. WASTE MANAGEMENT CENTER

The waste management center is for waste final treatment and disposal. This center is nearby the radioisotope production facilities.

2.5. INDEPENDENT REGULATORY BODY

Areas for nuclear regulatory supervision are reviewing and approving Safety Analysis Report (SAR) and issuing license.

3. DESCRIPTION OF RESEARCH CARRIED OUT AND ACHIEVEMENTS

The revised work plan for production of ⁹⁹Mo by neutron activation was implemented and the revised work plan for production of ⁹⁹Mo from fission of LEU plate targets as well.

3.1. PRODUCTIONS OF MO-99 BY NEUTRON ACTIVATION

The thermal analytical model has been developed by the CRP group for MoO_3 powder in quartz ampoule to be irradiated in Al can (see Fig. 1). Thermal hydraulic calculations have been performed as part of the SAR concerning safe irradiation of MoO_3 powder [1]. The irradiation position of MoO_3 powder ampoule and final quartz ampoule geometry were defined based on these calculations.

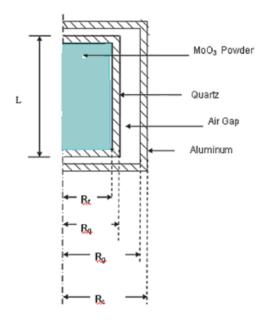


FIG.1. Schematic diagram of MoO_3 powder ampoule in Al can [1].

Handling procedures and shielded container for transportation were developed with supervision of the CRP group. A specific training has been provided by the CRP group to the operators for loading in shielded container and safe transportation. The prepared SAR by the CRP group concerning the safe irradiation, handling and transportation was reviewed and approved by the ETRR-2 Safety Committee with notification to the regulatory body.

The processing procedures together with QA procedures were developed. The processing hot cell for ⁹⁹Mo production by neutron activation was approved by regulatory body. The production cell personnel were trained on handling and processing of ⁹⁹Mo to produce ^{99m}Tc and the cell is ready for commissioning in cooperation with Chinese experts.

3.2. PRODUCTION OF MO-99 FROM FISSION OF LEU PLATE TARGETS

The facilities were supplied by INVAP SE within the scope of a contract for the irradiation of LEU plate targets and production of ⁹⁹Mo. The CRP group reviewed the modification of ETRR-2 core with two irradiation boxes on the basis of the IAEA safety requirements for research reactors [2]. The revision included commissioning tests of the modified core and updating of licensing documents, in particular the SAR, the Operational Limits and Conditions (OLCs), and the operation and training manuals. Independent thermal hydraulics and nuclear design and core management analysis were performed by the CRP group as revision and verification using validated research reactor codes (PARET, WIMSD4, CITVAP, and MNCP5). The results show that irradiation of the LEU plate targets is kept within the operational limits and conditions (OLCs) [3, 4].

The CRP group participated in the ETRR-2 core and hot cells modification. Two fuel elements in the reactor core were replaced by two irradiation boxes for ⁹⁹Mo plate target irradiation. The cooling system is the same for the fuel elements and for the ⁹⁹Mo targets i.e. no special cooling system or irradiation loop are required. The new reactor core contains 27 fuel elements, one in-core Cobalt Irradiation Device (CID) and two in-core ⁹⁹Mo targets irradiation positions as shown in Fig. 2. Each irradiation box has two target holders that can accommodate 12 plates of LEU, each plate having 1.4 gm of ²³⁵U as shown in Fig. 3. The used plate targets (130 mm x 35 mm x 1.4 mm) are made by CNEA (Argentina). The design and manufacturing of these plates are similar to the plates irradiated in the RA-3 reactor (Argentina). The CRP group participated in the modified core characterization and flux measurements verifying the hydraulic and nuclear design. The necessary modifications in the hot cells were implemented and the route for transportation of the irradiated plate targets from the reactor to the production cells was defined.

Within the scope of the contract, specific training was provided at ETRR-2 for operators on LEU plate irradiation, handling, and transportation. Also, the training on the processing for ⁹⁹Mo production was provided in CNEA (Argentina).

Two IAEA expert missions were requested by EAEA to support our team in the review of the SAR. These expert missions arrived in Egypt on Aug 2009 and July 2010 to support the review of the ongoing activities and advise in safety related issues. One IAEA expert mission on technological aspects of ⁹⁹Mo and ^{99m}Tc production at ETRR-2 was conducted in Aug 2010.

The irradiation of the LEU plate targets at the ETRR-2 reactor was done during the period from 23 October 2011 to 19 November 2011 with the participation of CRP group. The irradiation plan was started by irradiation of 6 target plates for 40 hrs. The second irradiation cycle was an irradiation of 6 target plates for five days. As a third irradiation, 12 plates were irradiated for 3 days. The irradiated plates were transferred to the RPF for commissioning the ⁹⁹Mo production hot cells and ^{99m}Tc loading cell and for training. The ETRR-2 Safety Committee with participation of the CRP group provided review and assessment on the safety and irradiation optimization during the commissioning tests and irradiation of plate targets. The CRP group participated in quality management of commissioning including verification, review, audits, and treatment of non-conformances.

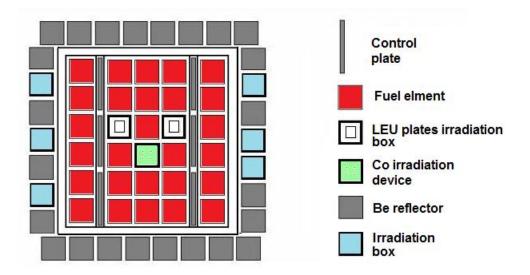


FIG. 2. Modified ETRR-2 core for irradiation of ⁹⁹Mo plate target.

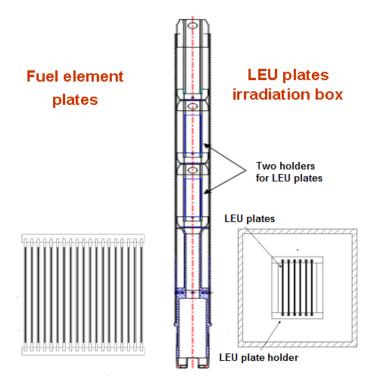


FIG. 3. Cross section view of fuel element and irradiation box at plates zone.

4. RESULTS

4.1. PRODUCTION OF MO-99 BY NEUTRON ACTIVATION

The thermo-hydraulic calculations for production of 99 Mo by activation method based on the analytical model developed for irradiation of MoO₃ powder in an ampoule of quartz is shown in Fig. 4 and Fig. 5. It is found that the maximum temperature at the center of ampoule is very low compared with the MoO₃ melting temperature (795°C). For more safety, it is proposed that the air gap region is replaced with aluminum foil to reduce the maximum temperature to avoid any uncertainties during irradiation [1]. As initial tests, irradiations of small amount of MoO₃ have been successfully irradiated.

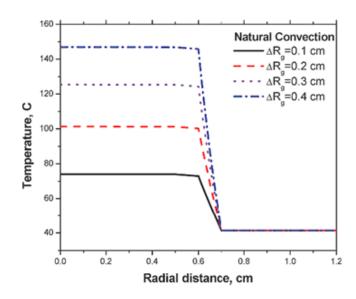


FIG. 4. The temperature distributions in Al can in case of natural convection [1].

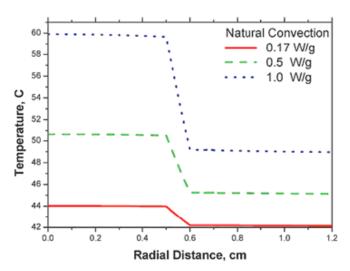


FIG. 5. The temperature distributions in case of natural cooling without air gap [1].

4.2. PRODUCTION OF MO-99 FROM FISSION OF LEU PLATE TARGETS

The thermal hydraulic design shows that with assembling two irradiation boxes in ETRR-2 core, the LEU plates can be irradiated at 22 MW with a total core flow rate of 1900 m³/h without compromising on reactor safety. The core LEU plates will have high safety margins of flow redistribution ratio (RDR) and departure from nucleate boiling ration (DNBR). Verification of the thermal hydraulic design is summarized in Table 1 [4]. The results of the commissioning tests demonstrate that the values of nuclear design parameters for the modified core are kept within the limits specified in the OLCs (see Table 2).

Measured neutron flux in the LEU plates for the six plates in the four possible positions (1, 2, 3 and 4) is show in Fig. 6 and Fig. 7 [4]. The redial flux distribution is shown as well as the axial measured flux relative to the calculated flux $(2.0 \times 10^{14} \text{ n/cm}^2 \text{ s})$.

The production of 3 batches of 99 Mo yielded 205, 340, and 523 Ci, with irradiations at the ETRR-2 of about 2, 3, and 5 days, respectively and 10 hrs cooling. The rated batch is of 500 Ci and the facility is built to produce 1000 Ci a week. One batch for training yielded 133 Ci for 3 days irradiation and 130 hrs cooling. Additionally, start-up and testing batches were produced for the loading of 99m Tc generators.

Parameter	Lower holder		Upper holder	
	PARET	TERMIC	PARET	TERMIC
	(CRP)	(INVAP)	(CRP)	(INVAP)
Maximum clad temperature	112 (107.4) °C	108 °C	116.1 (111.3)	113°C
T_w			°C	
Outlet temperature, T _{out}	49.7 °C	49 °C	57.4 °C	56 °C
Saturation temperature, T _{sat}	119.0 °C			
Onset of nucleate boiling	132.9 °C			
temperature, T _{ONB}				
RDR	5.1	5	4.6	3.2
DNBR	3.1	2.7	2.9	2.6

TABLE 1. CALCULATIONS FOR LEU PLATE TARGETS IN IRRADIATION BOX

TABLE 2. THE RESULTS OF COMMISSIONING TESTS

Parameters	Acceptance value	Measured value	
Core pressure drop (dp) for Hydraulic	New core $dp > dp$ of old core	+ 3.95 %	
verification of cooling system			
Shutdown Margin (SM)	> 3000 pcm	9360 pcm	
SM with single failure	> 1000 pcm	5300 pcm	
Reactivity Safety Factor	≥ 1.5	2.51	
The reactivity worth for Mo Box with 12	\leq 1200 pcm	340 pcm	
plates			
Power Peaking Factor	\leq 3	2.6 ± 0.3 %	

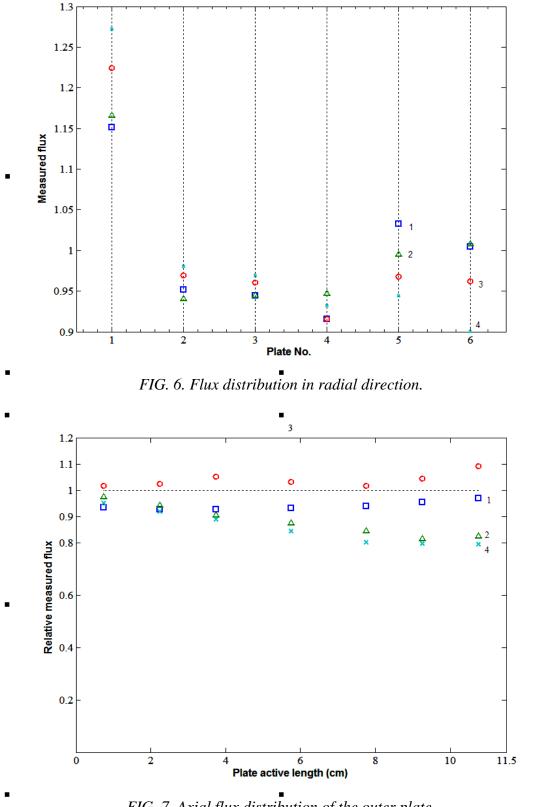


FIG. 7. Axial flux distribution of the outer plate.

5. CONCLUSIONS

As shown in the report, the EAEA developed the necessary production cells, irradiation facilities, and trained personnel staff to produce 99 Mo. The ETRR-2 can safely irradiate MoO₃

powder and LEU plate targets. The production cell for ⁹⁹Mo production from activation is ready for commissioning. A detailed safety analysis of ETRR-2 modified core has been performed to demonstrate safe irradiation and transportation of LEU plate targets. With the participation of CNEA- INVAP (Argentina) and EAEA (Egypt), the work towards the start-up of ⁹⁹Mo production from LEU plate targets has been successfully completed.

The CRP revised work plans for the production of ⁹⁹Mo from either activation of MoO₃ or LEU plate targets have been implemented.

REFERENCES

[1] ABD EL-HADY, A., ABOU EL-MAATY, T, Thermal analysis model for MoO_3 ampoules irradiated in a high neutron flux, kerntechnik 74 1-2 (2009) 47.

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