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STATUS UPDATE ON CONVERSION TO LEU BASED ^{99}Mo PRODUCTION IN SOUTH AFRICA

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ABSTRACT

The SAFARI-1 research reactor and NTP ^{99}Mo production facilities at Pelindaba continue to produce and distribute significant quantities of ^{99}Mo for the world nuclear medicine market. Over a number of years a significant effort has been made in the conversion of the molybdenum targets and ^{99}Mo production process from HEU to LEU.

History was made during the past 12 months with the first large commercial sized batch of FDA approved ^{99}Mo produced from LEU being shipped to the United States. This was the culmination of many years of work by various people and organisations.

CERCA, the French manufacturer of Research Reactor fuel elements and fuel target plates, continued with the development and refining of the target manufacture process and significant progress was made during the past year.

The emphasis has now turned to the challenge of increasing user uptake of LEU based ^{99}Mo and the practical challenges of production scheduling to allow both HEU and LEU production of ^{99}Mo as customers make their transition on different time lines.

This paper first looks back on the experiences of the past 12 months and then looks forward to the challenges being faced into the future.

1. Introduction

The SAFARI-1 research reactor is owned by the South African Nuclear Energy Corporation (Necsa). It was commissioned in 1965 and has a designed thermal power of 20 MW. Since commissioning, it has operated with an exemplary safety record. Commercial and research programs at the reactor are supported with an extensive infrastructure, ranging from theoretical reactor physics, radiochemistry and radio-analytical groups, a fuel and target fabrication plant, hot cell facilities for the production of medical and industrial isotopes, a pipe storage facility for the interim dry storage of spent fuel and a disposal site for low and intermediate radioactive waste. For the past ten years SAFARI-1 has typically operated at 20MW between 300 and 305 days per year for isotope production.

NTP Radioisotopes (Pty) Ltd, a limited liability company, is a wholly owned subsidiary company of Necsa. Its primary focus is the production and distribution of various radiochemicals to both the medical and industrial sectors. NTP also produces various radiopharmaceuticals such as its locally developed NovaTec-P ^{99m}Tc generator, ^{18}F based PET products and ^{131}I capsules and solutions.

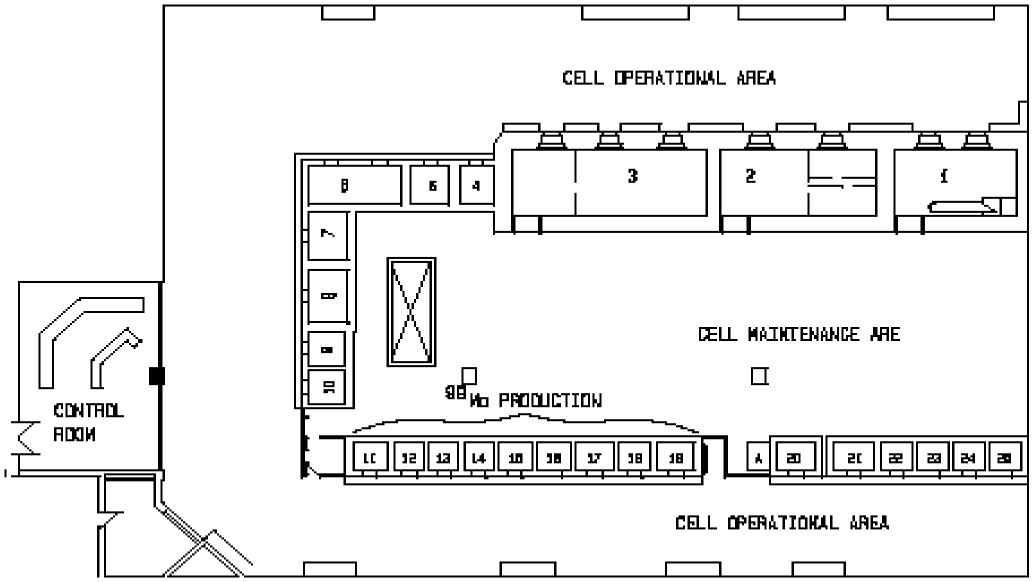
AREVA CERCA continued with the development of LEU dispersion targets to ever more stringent specifications. The co-operation agreement between Necsa and AREVA CERCA regarding the establishment of a fuel and target plant in South Africa based on dispersion technology continued [1].

Necsa and NTP have supported the principles of the Reduced Enrichment for Research and Test Reactor (RERTR) Programme for many years and have actively worked towards converting both SAFARI-1 to LEU fuel and the ^{99}Mo production process to LEU targets within the technical and commercial constraints facing it[2]. It should also be noted that despite the recent international crises with regards to ^{99}Mo production and supply, NTP continued to develop and industrialize its LEU based ^{99}Mo production process while ensuring the development programme did not influence routine ^{99}Mo supply.

During the past 12 months, significant milestones were achieved and these are outlined in this paper. In particular, the first ever commercial sized batch of FDA approved LEU produced Mo-99 was produced and shipped to the United States for patient use in December 2010.

2. Facility Description

NTP's radiochemical production facility consists of 3 large concrete shielded hot cells and 22 smaller lead shielded hot cells (See Figure 1). The extensive infrastructure also includes liquid waste handling and intermediate storage facilities for various activity levels; four ventilation and filtration systems (and additional specialized systems for the dissolver cells); solid waste handling and other support facilities. The facilities were originally used for the post irradiation examination of power reactor fuel but were substantially modified in the early 1990's to accommodate ^{99}Mo and other radiochemical production.



MOLYBDENUM PRODUCTION:- CELLS 15 TO 19
MOLYBDENUM PRODUCTION:- CELLS 11 TO 15

Figure 1: Schematic Layout of Hot Cell Facility

The ^{99}Mo production process was developed in the late 1980's and early 1990's and the first ^{99}Mo commercially exported in 1994. The plant was upgraded and scaled-up further based on the operational experience gained from the early years of operation.

The dissolution, extraction, purification and dispensing operations are spread over various hot cells in order to minimise the risk of cross contamination and provide an uncluttered and spacious environment to perform each step of the process. Two types of hot cells are used in a production line i.e. a special dissolver cell containing additional shielding, specialized ventilation and liquid waste systems in order to handle the highly radioactive target plates, and four standard cells housing the rest of the production processes. Currently the facility has two separate production lines for ^{99}Mo production and has dual dissolvers in each line.

The radiochemical production facility is operated for 51 out of every 52 weeks. This high level of operability is due to the availability of two production lines and extensive maintenance and upgrade programmes.

3. Conversion of ^{99}Mo Production Process

NTP has been producing ^{99}Mo from 45% enriched uranium for over 17 years and consequently has a wealth of experience of its process and plant. It was this experience base that was used to expedite the project to convert its production process to LEU.

In order to minimize the changes required to the target, irradiation, handling and processing, a low enriched uranium aluminium dispersion target was selected, the details of which are given in the table below.

Table 1: Target Plate Specifications

Parameter	**LEU**	**HEU**
Meat	Dispersion	Alloy
Enrichment	19.75%	45.0%
Uranium density ($g.cm^{-3}$)	2.75	1.42
Dimensions (mm)	200/50/1.66	200/50/1.66
Cladding	Alloy	Pure aluminium

Table 1: Target Plate Specifications

From 2008, numerous cold and hot development runs were performed. This was followed by formal process validation runs both for NTP as the producer of LEU ^{99}Mo and the ^{99m}Tc generator manufacturers. The time line of events is summarized in the table below.

Table 2: Timeline of events for ^{99}Mo production Process conversion

Date	**Event**
2007	Theoretical Feasibility Study Performed
2008	Cold and Depleted Uranium Experiments Performed
October 2009	Nuclear Regulator Approval Received for Hot Test Phase Hot Test Phase Commenced
March/April 2010	Process Validation Runs Performed
June 2010	Submission to Nuclear Regulator for Routine LEU ^{99}Mo Production Submission to Medical Regulators Commenced
July 2010	Customer Tests and Validation Runs Commenced
September 2010	South African Nuclear Regulator Approval Received for Routine LEU ^{99}Mo Production
November 2010	US FDA approves LEU ^{99}Mo for a customer in the US
December 2010	First large scale commercial FDA approved batch of LEU ^{99}Mo produced and shipped to US for patient use
June 2011	Routine commercial supply of LEU ^{99}Mo commenced to some customers

After the completion of the development work to optimize the ^{99}Mo extraction efficiencies, the yields obtained from the LEU ^{99}Mo production runs are equivalent to the HEU ^{99}Mo production runs. It must, however, be pointed out that the batch sizes have decreased by approximately 10% due to the ^{235}U loading in the LEU targets being lower. This is as a result of uranium density restrictions in the dispersed target.

The LEU ^{99}Mo produced continues to conform to the British and European pharmacopeias requirements.

The figure below indicates the distribution of LEU ^{99}Mo production runs performed in six monthly periods over the past 2 years. Other than the initial developmental runs performed in the latter half of 2009, there has been a steady increase in the number of LEU production runs performed during each of the six month periods over the past 18 months.

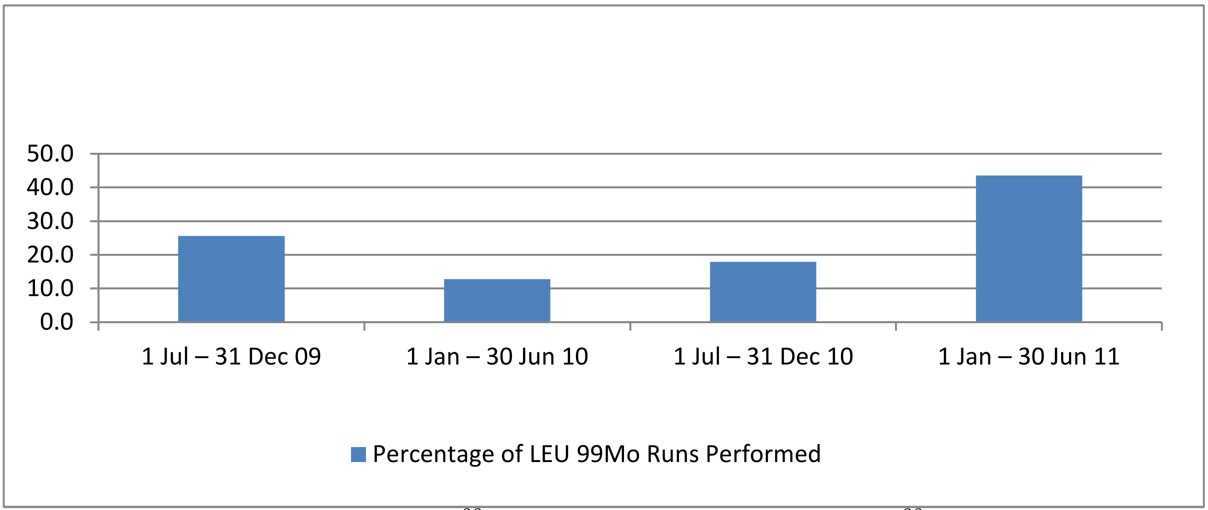


Figure 2: Percentage of LEU ^{99}Mo Runs Relative to Total LEU ^{99}Mo Runs

4. Manufacture of LEU dispersed targets

To support the conversion to LEU ^{99}Mo, the development of a new target plate – UAl_2 dispersion target – has been carried out and AREVA CERCA has met the challenge to manufacture this product on an industrial scale over a short period of time.

- AREVA CERCA manufacturing experience

AREVA CERCA has been successfully manufacturing and supplying research and material test reactor fuel assemblies for more than fifty years and has gained significant experience in this field. This experience covers a wide range of products, in terms of geometries (flat or rolled plates, tubular or ring-shaped elements) as well as enrichments (HEU, LEU) and fully satisfies the technical and scientific needs of customers demanding quality and safety.

Since 1960, AREVA CERCA has manufactured over 450 000 fuel plates, about 20 000 fuel elements of 70 designs, delivered to 40 research reactors in 20 countries. Among this production, about 5 000 are U_3Si_2 fuel elements, distributed to worldwide customers.

Besides the routine industrial manufacturing of fuel elements, AREVA CERCA is involved in several R&D programs, such as monolithic LEU targets and dispersive or monolithic UMo fuel development. These programs are run in collaboration with international partners and enable AREVA CERCA to increase its competences and knowledge of manufacturing processes.

- Dispersed LEU targets: status of industrial scale

In addition to the fuel manufacture and leveraging benefit from its knowledge in manufacturing, AREVA CERCA has been working for the past 4 years on the production of UAl_2 fuel targets. The first batches were produced and delivered to South Africa in 2008 and since then, several thousands of dispersed target plates have been manufactured and distributed.

Several steps of process optimization have been necessary to produce LEU dispersed targets and the manufacturing processes have been refined to comply with more stringent specifications which developed due to the experience gained in producing LEU ^{99}Mo.

Today, AREVA CERCA is manufacturing dispersed LEU targets in a reliable and industrial manner. Nevertheless, it has to be pointed out that this product is complex to manufacture and processes have to be mastered to achieve reliable supplies on an industrial scale.

The good cooperation between NTP and AREVA CERCA has enabled both parties to make excellent progress in understanding the relation between the quality of the targets and the final ^{99}Mo produced. The commitment from both organizations resulted in this development of an LEU dispersed target taking place over a short period of time.

5. Challenges

Putting the technical issues aside, there are various other challenges which have to be addressed in order to convert from HEU to LEU ^{99}Mo production. Some of these are mentioned below.

Customer Appetite
It is a significant, costly and time consuming exercise for a ^{99m}Tc generator producer to qualify themselves for LEU ^{99}Mo. Although customers are generally prepared to discuss the possibility of converting to LEU ^{99}Mo, there is a mixed response regarding the time lines of performing the required validation runs. Currently the benefit to the customers is minimal if any benefit at all.

Medical Regulators
Medical regulators require that a new Drug Master File (DMF) be prepared and submitted for LEU ^{99}Mo. This DMF together with the validation data provided by the ^{99m}Tc generator producers is then put through a formal review process. The challenge is that, not only does this take time, but it has to be done by the regulator of each country in which the ^{99m}Tc generators are sold. Only after approval is obtained from each of the regulators can the generator producer start manufacturing and distribution their generators with LEU ^{99}Mo.

Logistics
Due to the current routine production and supply of HEU ^{99}Mo and the requirement to minimize the impact of LEU ^{99}Mo production runs on current supply, production planning is becoming increasingly complex. Customers have rigid requirements regarding their ^{99}Mo supply due to downstream effects. As a result, in some circumstances, it is impossible to produce LEU ^{99}Mo on the day and at the quantities which the customer requires due to the detrimental impact on the HEU ^{99}Mo.

Also due to cGMP principles, complexities exist with regards to ensuring that the HEU and LEU production runs are separated to prevent any cross contamination.

^{131}I Production
Some ^{99}Mo producers also produce fission ^{131}I from the same targets used for the production of ^{99}Mo. The entire process which has to be gone through for the qualification of LEU ^{99}Mo

also has to be performed for LEU produced ^{131}I. The logistical challenges in this are also significant.

Waste Storage

Significant efforts and expenditure are currently being made by NTP to ensure that the storage capacity for the uranium residue containers is sufficient. The current facility was not designed to accommodate the additional mass and resulting increase in the number of uranium canisters. A hot cell and related transfer facilities are being designed and built to address this need.

Manufacture

The challenge is to increase uranium loading in the target in order to increase ^{99}Mo production capacity. This challenge, if achievable, will be the result of a joint effort, based on the investigation of manufacturing processes and product specification.

6. Conclusions

Significant progress has been made over the past 12 months both with regards to the target manufacture and the ^{99}Mo production runs. The number of LEU ^{99}Mo production runs continues to increase steadily with the associated experience base continuing to grow.

Some customers have completed their validation runs and obtained the necessary regulatory approval. This has resulted in routine commercial supply of LEU based ^{99}Mo commencing to some customers.

NTP continues to encourage other customers to proceed with their required validation runs and to submit the necessary documentation to the medical regulators. LEU ^{99}Mo is made available for these test and validation runs.

The challenges will continue to be addressed and solutions sought. In particular, the challenge of incorporating increasing numbers of LEU runs amongst the HEU runs, while minimizing the impact on routine HEU ^{99}Mo supply, is receiving attention.

NTP remains committed to completing the conversion of its ^{99}Mo process and switching over to LEU production but will do so in a responsible manner and will not place undue risk on the security of supply of approved ^{99}Mo.

6. References

[1] R.W. Jamie and A. Kocher, "Establishing a LEU MTR Fuel Manufacturing Facility in South Africa", Proceedings of the 14th International Topical Meeting on Research Reactor Fuel Management, Marrakech, Morocco, 21-25 March 2010.

[2] G. Ball, "Status of Conversion of the South African SAFARI-1 Reactor and ^{99}Mo Production Process to Low Enriched Uranium", Proceedings of the XXII International Meeting on Reduced Enrichment for Research and Test Reactors, Lisbon, Portugal, 10-14 October 2010.