

**Mo-99 2013 TOPICAL MEETING ON  
MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT**

**April 1-5, 2013  
Embassy Suites Downtown - Lakeshore  
Chicago, Illinois**

**Industrialization of LEU 99-Mo Target  
Production in AREVA-CERCA**

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**ABSTRACT**

This paper reviews the progress made during the industrialisation of the LEU  $^{99}\text{Mo}$  target. Started from prototype in 2007, the target is now produced on industrial scale. The road has not been easy and many difficulties were addressed. Meanwhile, following target development and qualification performed at AREVA-CERCA, LEU  $^{99}\text{Mo}$  targets were delivered allowing work to start on an appropriate irradiation and processing qualification program and validation of the end use of  $^{99\text{m}}\text{Tc}$ .

The historical developments and methodology from the preliminary R&D program up to Six Sigma and Lean manufacturing techniques which led to the industrial production are presented. An update on LEU  $^{99}\text{Mo}$  target production and the AREVA-CERCA view on manufacturing, the qualification pathway and industrialization is provided.

**1. Introduction**

AREVA-CERCA manufactures and supplies fuel elements and  $^{99}\text{Mo}$  targets worldwide respectively to research reactors and Mo99 processors.

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. Its experience covers a wide range of products, in terms of geometries (flat, curved, circular, ring-shaped elements) as well as enrichments and fully satisfies customers requirements in terms of high quality and safety.

Since 1970 AREVA-CERCA has manufactured over 50 000  $^{99}\text{Mo}$  targets. Historically uranium-aluminium dispersed HEU targets were produced first. Supporting the conversion from HEU to LEU, AREVA-CERCA has developed a manufacturing process to produce new LEU targets [1] from an existing concept [2, 3]. An R&D program started in 2007 to produce uranium-

aluminium dispersed LEU targets [1]. The first batches were produced and delivered in 2008 and since then several thousand LEU targets have been manufactured and distributed reliably to South Africa and Australia. Today, HEU and LEU targets are routinely produced and delivered.

## 2. Technological challenges

The  $^{235}\text{U}$  content of the target has a direct impact on the quantity of  $^{99}\text{Mo}$  finally obtained. The side effect of LEU is to decrease the  $^{235}\text{U}$  content. In order to keep the same final  $^{99}\text{Mo}$  yield the loss of  $^{235}\text{U}$  per target must be compensated. With the same uranium alloy and the same target geometry the loss of  $^{235}\text{U}$  is not balanced. The loss of  $^{235}\text{U}$  is compensated by adjusting the proportion of the higher density uranium alloy and/or an optimised target geometry. In this chapter, we will discuss the consequences of such changes and the associated technical challenges.

- High density uranium alloy:

The LEU targets are made of  $\text{UAl}_2$  and Al powder mixtures at the beginning of the manufacturing process. The density of  $\text{UAl}_2$  is  $8.14 \text{ g/cm}^3$ . During the manufacturing process, heat treatments are applied to transform  $\text{UAl}_2$  by activating the following solid state chemical reaction:  $\text{UAl}_2 + \text{Al} \rightarrow \text{UAl}_3 + \text{UAl}_4$ . At the end of the manufacturing process,  $\text{UAl}_2$  is transformed into  $\text{UAl}_3$  and  $\text{UAl}_4$ .

The advantage of this process is to obtain uranium alloys with density close to  $\text{UAl}_2$ . However, (1) the solid state chemical reaction  $\text{UAl}_2 + \text{Al} \rightarrow \text{UAl}_3 + \text{UAl}_4$ , (2) the U-Al metallurgical properties and (3) the mechanical constraint due to the meat volume changes must be controlled. If the production processes are not well controlled, then quality specification requirements may not be respected such as clad/core debonds or minimum cladding thickness.

- Uranium volume fraction in the  $\text{UAl}_2$  target meat:

The dispersed fuel and target meats are made of a mixture of soft powder (Al) and hard powder ( $\text{U}_3\text{Si}_2$ ,  $\text{UAl}_x$  or  $\text{UAl}_2$ ). The  $\text{U}_3\text{Si}_2$  and  $\text{UAl}_x$  fuels have been produced industrially for several decades. The industrial limits of the volume fraction of the hard powder in the meat is about 45 % for  $\text{U}_3\text{Si}_2$ , corresponding to  $4.8 \text{ gU/cm}^3$ . Since the rolling behaviour of  $\text{UAl}_2$  is similar to  $\text{U}_3\text{Si}_2$ , a limit of  $\text{UAl}_2$  uranium density can be defined within the range of 2-3  $\text{gU/cm}^3$  -equivalent specification-. The value depends on the cladding aluminium alloy, target geometry and quality specification requirements. AREVA-CERCA feedback has evidenced that the density should be limited to around  $2.5 \text{ gU/cm}^3$  in order to guarantee reliable target delivery and safe target use.

- Target meat geometry:

The high uranium density is not sufficient to compensate the lower enrichment. Increasing the geometry of the target uranium meat is a natural way to increase the  $^{235}\text{U}$  loads. However, this solution impacts the irradiation conditions and need to be addressed with reactor operator and  $^{99}\text{Mo}$  processor for safety reasons. All geometrical modifications should be then analysed according to the robustness of the reactors and process or against the new dimensions making the production cost compatible with market demand.

### 3. Industrial challenges

In parallel to the R&D, AREVA-CERCA addressed the industrial transition needed to meet the increasing demand for LEU targets. This transition has been addressed using a conventional quality program performed on site together with the implementation of specific well-designed quality tools selected to obtain rapid improvements.

Close cooperation between AREVA-CERCA and its customers (i.e. NTP and/or ANSTO) was required in order to achieve a constant and high level of quality. This enabled the parties to make excellent progress in understanding the relation between the target quality, irradiation aspect and final  $^{99}\text{Mo}$  quality when produced. The manufacturing processes have been optimised in several steps and refined to comply with more stringent specifications developed during the production of LEU  $^{99}\text{Mo}$ .

- Low tungsten:

An example of a new and more stringent specification is the low tungsten content to be met.

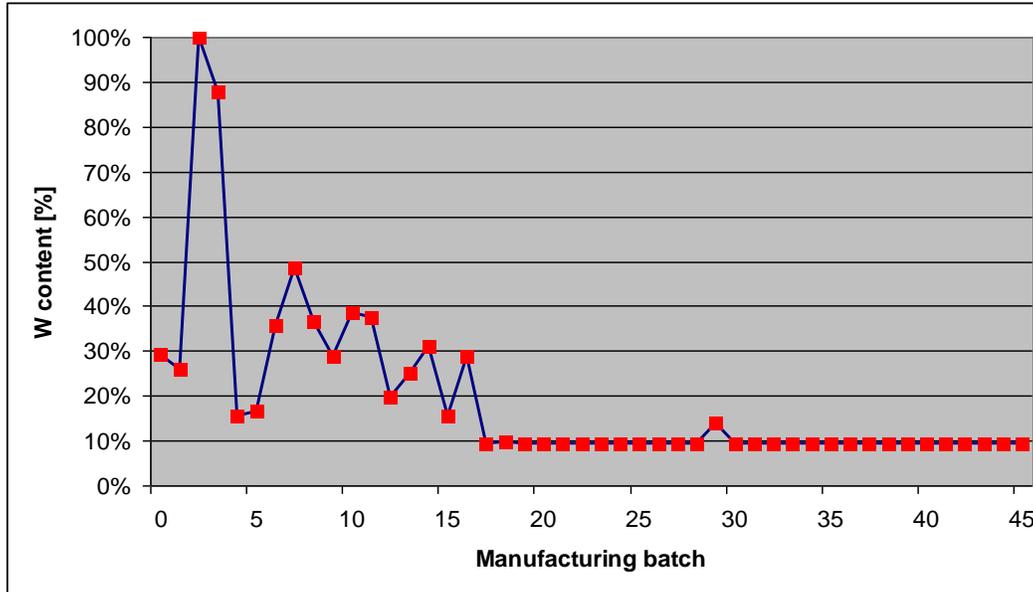
The industrial methodologies deployed were *Six Sigma* [4] and *Lean manufacturing* [5]. *Six Sigma* seeks to improve the outputs process quality by identifying and removing or reducing the source of the defect and minimizing the production variability of the processes considered. *Lean manufacturing* is centred on preserving value for customer with smart adaptation of the processes. Several tools are implemented to reach this goal, for example *Value Stream Mapping*, *5 S* and *Single-Minute Exchange of Die*.

In 2010,  $^{187}\text{W}$  impurities were reported during the production of  $^{99}\text{Mo}$  and we found that AREVA-CERCA LEU targets had a concentration level of a few hundred ppm, which was defined as damageable for the quality of the  $^{99}\text{Mo}$  [6].

The *Six Sigma* methodology is composed of five phases, with the acronym DMAIC (Define, Measure, Analyse, Improve, Control). The goal *definition* consistent with the customer's demands was to reduce the tungsten content on the raw target material. A tungsten measurement method was developed first. It can detect the whether or not tungsten is present with a detection level of a few tens of ppm. In the  $\text{UAl}_2$  target manufacturing process more than 100 variables, with potential impact on product properties, were identified. The most critical variables were identified and *measured* during several months. These data were *analysed* to investigate and establish the cause-and-effect relationships. The process *improvements* were based upon this data analysis and standard work to create a new state process. Several improvement steps were necessary to decrease the tungsten impurity to an acceptable level. Finally a new state process was established and a suitable *controlled* plan put into place to ensure tungsten conformity. Statistical process controls were implemented and since then the process has been continuously monitored.

Figure 1 shows the change in tungsten content during the project, showing the benefits of the processing improvements and successful use of this *Six Sigma* project. Today the tungsten content of AREVA-CERCA LEU targets complies with the customer's expectation.

Over a short time period, 12 months, AREVA-CERCA has been mobilized to comply with the customer's expectations. During this period the delivery dates were met and all the LEU targets were supplied to the customer in due quantity and quality.



**Figure 1:** Tungsten content variation in LEU target Uranium alloy (in percent)

- Target geometry

Another industrial challenge we faced was to combine geometry requirements such as the meat thickness that the <sup>99</sup>Mo producer would like to be as thick as possible with a high quality and reliable supply.

The target design - which belongs to the customer - is defined taking into account the constraints of all parties involved (manufacturer, irradiation reactor and <sup>99</sup>Mo processor).

The target geometry may be inside or outside the AREVA-CERCA validated fuel meat range.

If the selected geometry is inside the AREVA-CERCA production range, the validation road is short from design to routine production. A target Depleted Uranium (DU) batch is usually produced in order to validate the production process and expected yield.

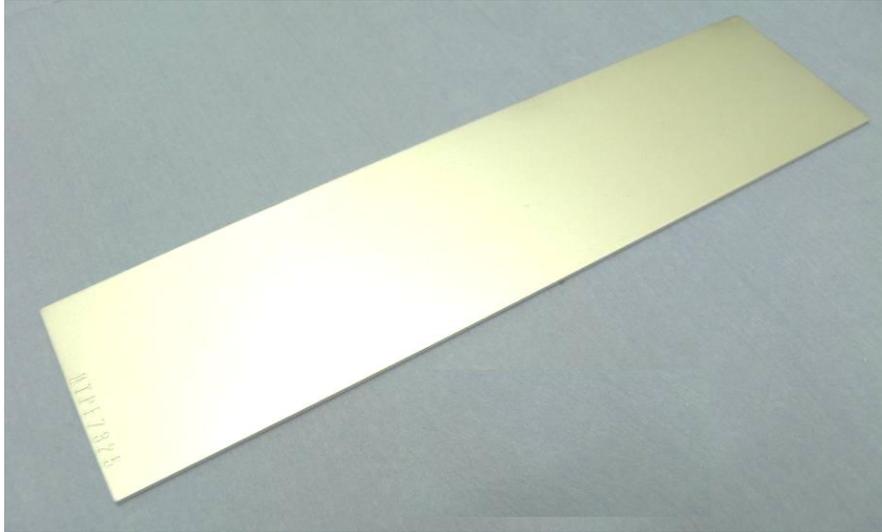
If the selected geometry is outside the AREVA-CERCA production range, the road to reach a routine delivery is longer. A preliminary feasibility program must be added. This feasibility is requested to define the appropriate equilibrium between risk, schedule and technological challenges.

#### 4. Industrial production status

AREVA-CERCA currently manufactures LEU targets routinely, reliably and on industrial scale. Each year, thousands of LEU targets are supplied worldwide from the AREVA-CERCA production lines with high quality levels and on-time delivery.

AREVA-CERCA routinely supplies UAl<sub>2</sub> targets to NTP Radioisotopes (Pty) Ltd, a NECSA subsidiary [7, 8], and ANSTO Radiopharmaceuticals [9, 10]. The excellent cooperation between AREVA-CERCA and its customers has led to a better understanding of the relation between target quality, safe irradiation and final <sup>99</sup>Mo quality. To date, all AREVA-CERCA targets have been safely irradiated and high quality <sup>99</sup>Mo processed.

AREVA-CERCA has started LEU conversion programs with NRG/COVIDIEN and IRE. An  $UAl_2$  LEU target has been selected as technical option. The qualification programs are defined and run as scheduled.



**Figure 2:**  $UAl_2$  LEU target picture

## 5. Security of supply

Due to its strong position on the research reactor fuel and  $^{99}Mo$  target market, AREVA-CERCA has always been committed to being a responsible and responsive player and is fully aware of the public health implication of the products it supplies.

AREVA is committed to supporting its customers and assisting them in all fields closely related to  $^{99}Mo$  production.

AREVA manages the operational risks related to the manufacture of targets by upgrading its manufacturing facility and controlling its product quality.

Sustainable procurement of uranium: since the targets are made from uranium compounds, their production depends on the supply of uranium. Procurement of enriched uranium is strictly regulated by International policy, exported under stringent conditions and only through governmental agreements. AREVA helps ensure on-time delivery by providing administrative and logistic support, and offers options to accommodate even critical situations. A sufficient uranium stockpile is necessary.

- Safe transport: transportation is a transverse activity which is performed first for uranium which needs to be imported to France, for fresh targets to be transported to the appointed research reactor, and finally for the irradiated targets to be transported to the  $^{99}Mo$  processing. The transport risk is mainly related to the on-time disposal of suitable, dedicated and licensed containers. Mitigation of this risk can be evaluated with the regulators involved, experts on the cask licensing activity, reactor operators and  $^{99}Mo$  producers. Moreover, AREVA

manages transport insurance, license application and other administrative issues in a successfully and timely manner.

- Available manufacturing facility: targets are manufactured in a fuel production line in a nuclear installation which is strictly controlled and regulated by independent Safety Authorities. This safety culture and the associated organization mitigate operational risks related to unexpected major events which could result in production shutdown. The first safety and security principle we apply is based on “defence in depth” where more than one barrier of defence is requested for each identified risk - global and/or local - and, if the situation occurs, it should be brought under control by dedicated means and/or with an acceptable time where the “effect” should then be confined in a specific space.

The AREVA-CERCA facility is assessed every 10 years by an independent group of experts mandated by the French Nuclear Authority (ASN) [11]. The ASN assesses the facility routinely several times a year and makes recommendations which must be strictly followed. Safety investments are made accordingly and safety improvements are mandatory in order to renew the license and operate the plant.

In order to ensure target security of supply, suppliers must commit to the business on a long term basis keeping the technical ability to upgrade the facility as requested by regulators and therefore being ready to make significant investments on a regular basis over many years. Industrial fuel and/or target suppliers need confidence in the market to make this financial commitment.

- Product quality: another operational risk is related to the quality of the products: any quality issue that occurs may result in postponement of the delivery while waiting for dedicated measures to be implemented. This risk for HEU targets is extremely low since the knowledge of this product, stated in the specifications, is extensive and has been shared for a long time between the parties involved - manufacturer, reactor and  $^{99}\text{Mo}$  producer.

The conversion program, defined as the entire action plan to be implemented in order to obtain the appropriate license / validation for authorizing the use of LEU target is carried out by a joint and commercial approach between  $^{99}\text{Mo}$  producers, reactors and target supplier.

The transition from HEU to LEU targets includes, by definition, a certain degree of uncertainty defined and bounded by the strategic and technical options selected by the  $^{99}\text{Mo}$  producers and/or the dedicated operator in charge of the conversion program. Some of the technical uncertainties, related to the targets manufacturing, were reduced through R&D investments.

Long production series over many years have enabled us to gain experience and develop the statistical approach necessary to implement the appropriate improvements and keep the process under control. The diversity of customers and specifications provide opportunities to explore process abilities and enrich our experience and knowledge. As a result, the efforts and planning to achieve production on industrial scale have been reduced.

Nevertheless, unexpected events – internal or external - may occur in the  $^{99}\text{Mo}$  supply chain. In order to secure the  $^{99}\text{Mo}$  supply and avoid any detrimental risk of shortage it is recommended to build an optimised stockpile of fuel targets.  $^{99}\text{Mo}$  producers should investigate this strategic approach, which is the only way of facing any risks arising from technical, transportation and uranium supply issues.

## **6. Continuous improvement and innovation**

AREVA-CERCA has an active R&D team which has been involved in the worldwide development of LEU targets for several decades. In the past, AREVA-CERCA has developed uranium nitride LEU dispersed targets [12]. Studies on uranium silicon high density fuels [13, 14] inspired uranium silicon LEU dispersed targets [15].  $UAl_2$  targets are continuously improved in order to satisfy our customers. Active developments are also under way on uranium metal LEU monolithic targets.

Uranium metal LEU monolithic targets have been developed by Argon National Laboratory (ANL) [16, 17] and manufacture has been demonstrated by AREVA CERCA [18, 19]. The processes and equipment are qualified.

The transition from tubular target design to flat design was originally proposed by AREVA-CERCA and MURR [18, 19]. Production of flat and/or semi-curved targets was demonstrated and is awaiting partnership for further developments and validation, i.e. irradiation validation test and/or Mo processing.

AREVA-CERCA is ready to investigate any LEU target option that customers can provide and/or that any international agencies want to develop.

## **7. Conclusion**

The production of LEU targets is more complex than the production of HEU targets. AREVA-CERCA has leveraged benefit from its experience gained thanks to customer partnerships.

AREVA-CERCA manufactures LEU targets routinely in a safe, industrial and reliable manner. Each year, thousands of LEU targets are supplied worldwide from the AREVA-CERCA production lines with high quality levels and respect of schedules. AREVA-CERCA is adapting its production infrastructure and organization to meet the global demand for LEU targets.

As key stakeholder in the  $^{99}\text{Mo}$  supply chain AREVA-CERCA is willing to participate in international LEU  $^{99}\text{Mo}$  conversion projects. AREVA-CERCA is continuously improving the existing LEU targets and looks forwards to new LEU target designs.

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