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Background and Projects on LEU based production technologies of Molybdenum 99 within INVAP

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ABSTRACT

INVAP has been developing, constructing, commissioning and turning over multipurpose research reactors and the associated production facilities for the past 35 years, all based on LEU. In particular and with the support of the Argentinean Atomic Energy Commission, a successful tailoring and dimensioning of LEU based production technologies of molybdenum 99 and other radioisotopes has been achieved in the cases of Australia and Egypt where unique powerful multipurpose research reactors where delivered on a turnkey mode. More recently a subcritical miniloop boosted by the RA 6 reactor core has been installed for the development of a separation process from an LEU aqueous solution. Achievements, problems and issues solved through above experience and, future programs and trends are presented in this paper. In the conclusions some suggestions for thought are also outlined.

1. Introduction

INVAP has been developing, engineering, constructing, commissioning and turning over multipurpose research reactors and the associated production facilities for the past 35 years all based on Low Enriched Uranium (LEU) and all with increasing performance. To achieve in the field, many factors have to be considered, but aside from the intrinsic capabilities within the Company, the other important factor is the history, tradition, infrastructure, skills and programs in the hands of the Argentinean Atomic Energy Commission (CNEA).

2. Background on Radioisotopes in Argentina

The Argentinean Atomic Energy Commission started to develop, produce and employ radioisotopes for nuclear medicine in the late 60s and early 70s making Argentina a pioneering country in the matter. In 1973, the first Radioisotope Plant was built in the Ezeiza Atomic Center (in the outskirts of Buenos Aires and attached to the RA3 research reactor) for the production of I-131. The plant made Argentina self - sufficient in those days. Early in those years too, the RA3 reactor started the irradiation of natural Molybdenum Oxide (MoO_3) to produce, by activation, the Mo99 used in the low activity generators of those days. The generator itself was developed in 1975 in the country. But by the middle of the 80s it was clear that the medical community, the extension of the country and the growing propulation needed higher activity generators and a process was adopted, developed and implemented to produce Mo99 from the fission of U235 (again employing the RA3 reactor and new facilities in the Radioisotope Production Plant). The operations started in April 1985 and again a local Tc-99m generator called GENTEC was developed. Up until 1998, the whole activity was carried out by CNEA until the generator portion of the business and their distribution was privatized. In all those years still employing Highly Enriched Uranium (HEU) targets irradiated in the RA3 reactor. Again, Argentina became part of a reduced group of countries in handling the associated technologies that except for the HEU itself was self – sufficient in the rest of the process.

In those two decades, Argentina, CNEA and the rest of the nuclear system always committed with nuclear non - proliferation principles, started to work on Low Enriched Uranium fuel concepts / solutions and became active participants of the Reduced Enrichment for Research and Test Reactors program (RERTR). By 2000 – 2001 the practical ban in accessing to HEU plus the economic crisis made impossible for the country to have an assured supply of radioisotopes. These facts forced the production system to convert the fission based processes from HEU to LEU totally in house and with the experience of the previous 25 years. By 2002 the new production system based on LEU was well developed, the production of radioisotopes was fully converted (without closing or stopping during the conversion) and the country was self - sufficient in the supply of Mo99 without the need to relying on imports (other than during reactor maintenance) and avoiding the expenditure of the scarce foreign currency needed for imports.

3. INVAP

Established in 1976, INVAP is a technology project oriented company specialized in the development of multi-disciplinary technological projects within the nuclear, space, industrial services, medicine, and information-technology fields. Following specific client requirements or first of a kind needs, INVAP provides services and products ranging from consultancy to turnkey facilities, essentially all of them first of a kind solutions. The company has the means to engage in every stage of a project cycle, namely feasibility studies, development, design, engineering, supply, manufacture, construction, commissioning, start-up, operation, and post-sale services.

INVAP sprang from within the scientific community, and is led by scientists with the old entrepreneurial approach. It has had only two CEOs throughout its history - both physicists -, the other top managers being either physicists or chemists. Most of INVAP's staff was trained in the field of applied research, and comes mostly from within the ranks of the Argentine Atomic Energy Commission (CNEA). This profile guarantees competitiveness towards the accomplishment of projects involving many disciplines, development tasks, and custom-design technologies.

Nonetheless, but in sync with its scientific profile, INVAP is a profit-based organization. The company shares are wholly owned by the Province of Río Negro. INVAP's only sources of income are the revenues from its contracts, in light of which it had to develop the managerial and commercial capabilities of its top managers since its inception. INVAP today has more than 900 highly qualified directly employed people and more than 300 indirectly working in related or subcontracted companies. Company total turnover is right now higher than 70 MUSD per year depending on the project load.

INVAP, as an arm of the strategic goals of the country in the technology field has devoted a strong effort on working not only within the Argentine market but also projecting the Argentine technology to foreign clients. INVAP works in close cooperation with the Argentinean R&D complex, especially with the Atomic Energy Commission (CNEA), the Aerospace Activities Commission (CONAE), the Ministry for Science & Technology, different recognized universities and laboratories and the satellite communications company AR-SAT. INVAP, in a project oriented mode bridges the gap between basic knowledge and the society needs through the engineering of applied technology solutions. Innovation and dimensioning to specific needs is key in this process. In the past 35 years therefore the basic knowledge developed by the nuclear scientific community has been converted into practical customized solutions in Argentina and abroad.

Through more than 35 years INVAP has built a reputation as prime contractor of complex technology-intensive projects deployed both locally and abroad. Within the US and Canada , INVAP is a subcontractor of and consultant to Westinghouse Nuclear, B&W Nuclear, Energy Solutions, Atomic Energy of Canada Limited (AECL) and a supplier of space satellites for the U.S. National Air and Space Administration –"NASA"-

Fig 1 – Evolution of INVAP presence in the research reactor market

The Nuclear Business Unit specializes in the delivery of customized turn-key nuclear projects. One of INVAP's most notorious achievements was the indigenous and autonomous development of the Uranium enrichment technology for Argentina (1983). INVAP has delivered many nuclear facilities and developed projects in areas such as: research reactors, demonstration reactors, uranium processing facilities, fuel manufacturing facilities, waste storage, services to nuclear power plants, extractive metallurgy, metals processing, nuclear medicine, radioisotope production plants, etc. In the past 25 years INVAP has designed, built and delivered 6 research reactors and associated facilities in Argentina and abroad as depicted in above figure. All those facilities based on LEU both the fuel or a as the raw material for production of radioisotopes and in doing so INVAP has demonstrated that HEU is not necessary for achieving higher performances in civilian uses.

INVAP quality assurance system is ISO 9001:2000 certified, and complies with IAEA SS 50-C/SG-Q, ASME NQA-1 (all the levels, all the requirements) and NRC 10 CFR Part 50 Appendix B requirements.

4. CNEA Experience in Radioisotopes

CNEA has been producing LEU base Mo-99 to supply the needs of the national market for ten years. Presently it is also supplying neighbouring countries such as Brazil (50% of its weekly needs) plus Uruguay, Paraguay and Chile. Mo-99 production is carried out in the Radioisotope Production Facility associated to the RA3 research reactor at the Ezeiza Atomic Centre.

The facility has two laboratories with four airtight cells each. Two cells for uranium plate Dissolution Process and two cells for Purification Process. There are two main cells and two auxiliary cells for Production and Transfer. Purification cells are connected to the auxiliary cell through transfer tunnels, with a movable carrier in each tunnel and pneumatically-activated doors on their ends. Airtight boxes are of stainless steel, with airtight-closure conduit connectors for the passage of gases, liquids, and electricity.

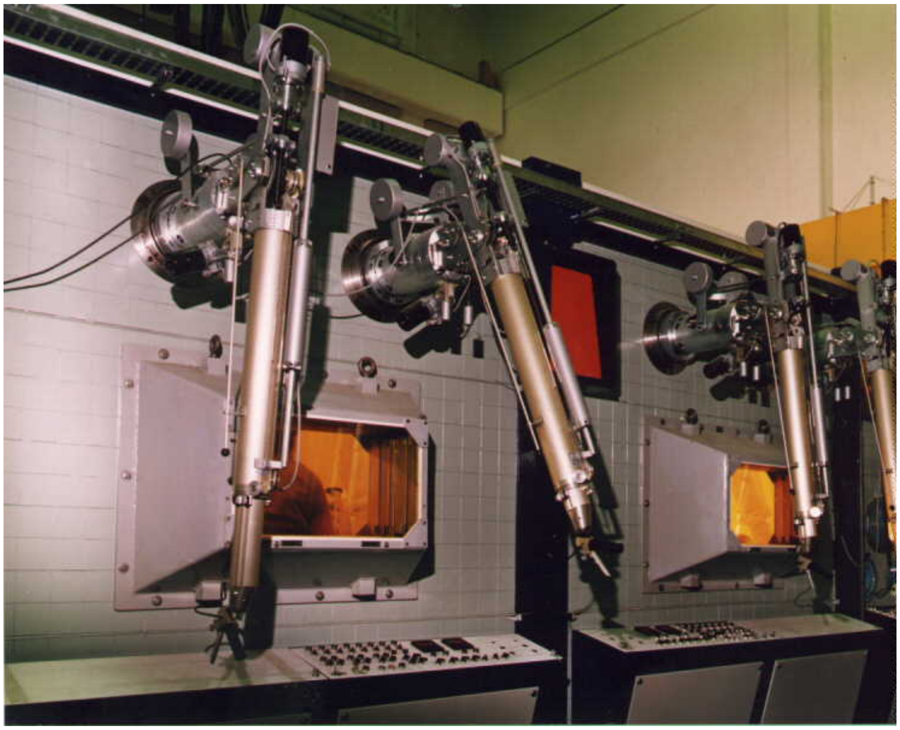

Fig 2 - Front View of the Process Cells

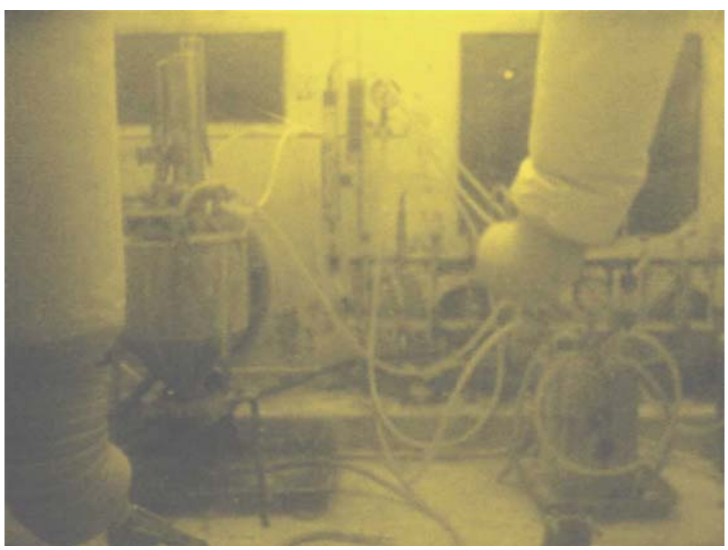

Fig 3 - Inner View of the Dissolution Cell

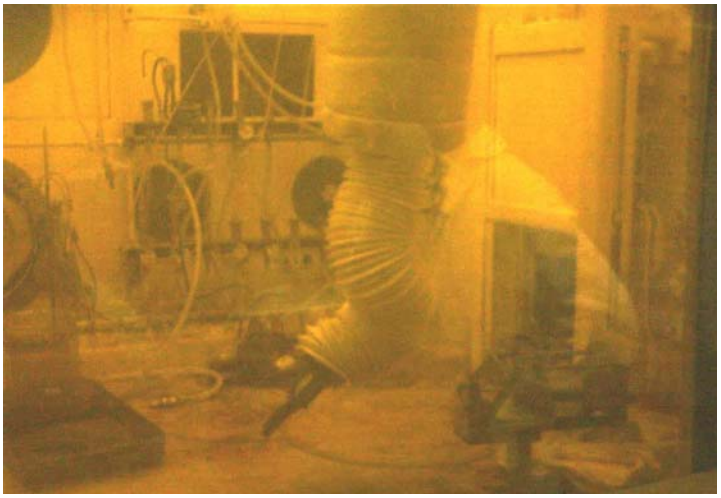

Fig 4 - Inner View of the Purification Cell

Fig 5 - Inner View of the Purification Cell

5. ECRI Facility for Manufacture of FE and Irradiation Targets

CNEA owns and operates the ECRI facility located at Constituyentes Atomic Centre. The plant manufactures MTR-type fuel elements for research reactors in Argentina and abroad, and irradiation targets to obtain Mo-99.

The fabrication facility is divided into four main areas, each with different zones provided with manufacture and control equipment and processes: Controlled Area for the manufacture of uranium-based semi-finished products, Fuel Plate Area for the manufacture of covers and fuel plates, Assembly Area, and Quality Control Area.

The experience of CNEA and INVAP in the development and production of fuel elements for research reactors and molybdenum targets is shown in the following table.

Reactor Country	Contract Year	Fuel Type	Density (g/cm3)	Quantity Supplied	Irradiation of Plate Targets
RA2, RA3, RA6 Argentina	1963-1987	MTR HEU	0.6	312 FE* 135 CFE*	-
RP10 Peru	1987	MTR LEU	3.1	23 FE 5 CFE	-
NUR Algeria	1989	MTR LEU	3.1	36 FE 9 CFE	-
RA3 conversion to LEU Argentina	1990	MTR LEU	3.1	27 FE 8 CFE	Yes
TRR2 Iran	1992	MTR LEU	3.1	66 FE 12 CFE	-
ETRR2 Egypt	1997	MTR Type 1** MTR Type 2**	3.1	30 FE	Yes
RA6 conversion to LEU Argentina	2009	MTR LEU	4.8	25 FE 6 CFE	-
OPAL Australia	2006	MTR LEU	4.8	63	Yes

FE: Standard fuel element
CFE: Control fuel element

MTR Type 1: 147 g U-235
MTR Type 2: 209 g U-235

CNEA developed and qualified HEU-based targets during the 1970s, manufacturing these targets for the Mo-99 production facility, over 3000 plates, for 20 years. CNEA replaced completely the HEU-Al alloy with the LEU targets in 2002, which were developed and qualified during 1999 and 2000.

The following pictures provide views of the target plates used for Mo-99 production in CNEA.

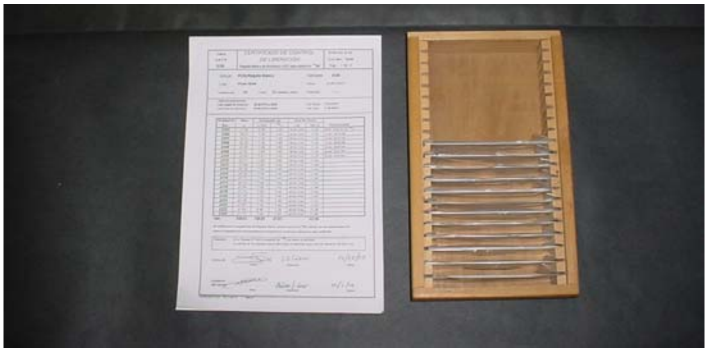

Fig 6 - Mo-99 Targets Quality Control

Fig 7 - Mo-99 Targets

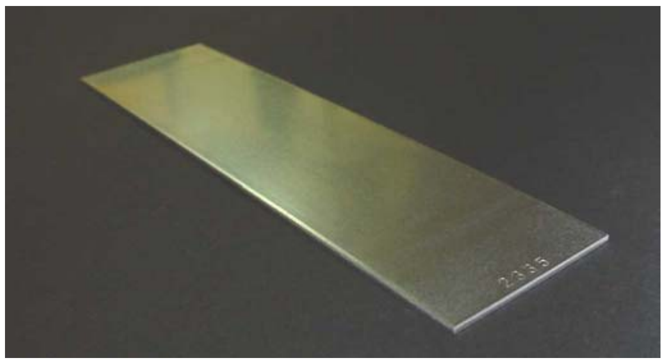

Fig 8 - Mo-99 Target Miniplate

6. Radioisotopes – Activities and Cycle

Between the raw material (in our case Uranium since we consider that the Molybdenum activation option still needs further work as a viable solution for present market needs) and the patient / human being the production chain can be divided into the following major steps.

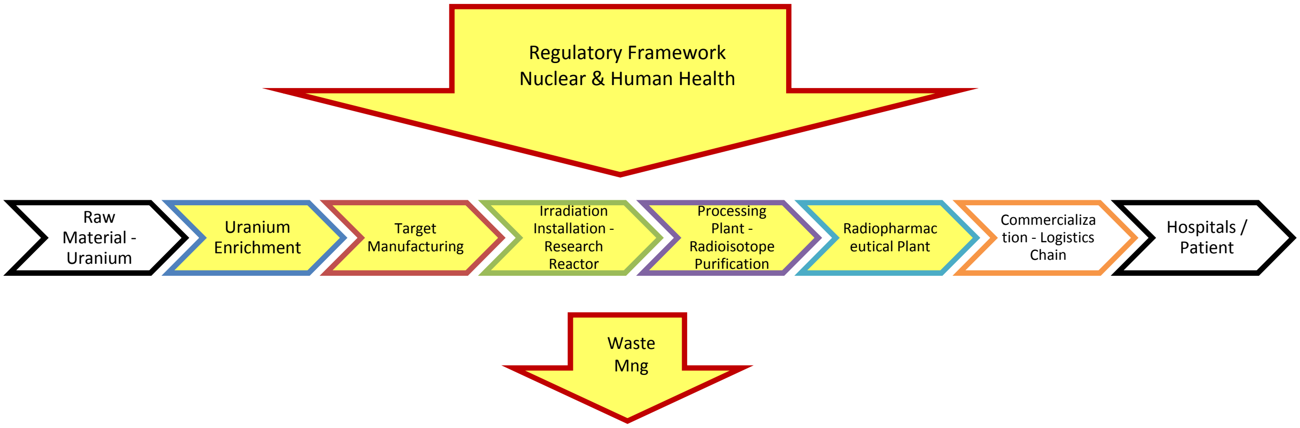

Fig. 9 – Radioisotope Production Activities and highlighted, INVAP areas of work

INVAP has expertise and dedicates part of its technical endeavors to all the steps except both ends: Commercialization – Logistics, the Patient and the Raw Material. In other words, INVAP does not deal and is not interested with the distribution, commercialization portion of the market.

INVAP as a technological company has focused on projects related essentially to the Irradiation Installation and the Processing and Purification Plant. Waste Management, an integral Regulatory support to our customers and target manufacturing in collaboration with CNEA are also important areas.

Since in some other conferences and papers it is well described the capabilities of the Company in the area of the Installations for Irradiation (namely and for our case multipurpose LEU high flux research reactors like OPAL in Australia and ETRR II in Egypt) hereby we will concentrate on the efforts and achievements related to Processing and Purification Installations.

7. INVAP Radioisotope Processing Plants – Large Scale Projects

In the last 12 years, two important and unique projects have been completed with success. One started 11 years ago in Egypt for the Atomic Energy Authority (AEA) as a part of providing such a country with the associated installations for a full use of the Egyptian Test and Research Reactor ETRR II of 22 MW thermal that we also built and is in operation since 1997. This project involved the supply on a turnkey mode of a Radioisotope Processing Plant (RPF) devoted to make Egypt self - sufficient in its needs for them.

The other significant achievement was in Australia where during the last stages of the OPAL project INVAP was asked by ANSTO to retrofit with a completely new process its existing infrastructure of operating hot cells in Lucas Heights.

The projects as can be seen, ultimately aimed to produce LEU based radioisotopes on a weekly significant scale basis are in their nature very different: one implying a completely new state of the art facility (Egypt), the other implying the adaptation, engineering and installation of a new process in existing and contaminated hot cells.

8. AEA Radioisotope Production Facility (RPF-Egypt)

INVAP supplied AEA (Atomic Energy Authority of Egypt) a turn-key facility (including Civil Works and Auxiliary Services) for production of radioisotopes for medicine, industry and research activities. The technology is based on a adequately sized version of the CNEA process in use in Argentina.

Fig. 10 – Radioisotope Production Facility (RPF) inside Inshas Atomic Center - Egypt

The facility operates with targets irradiated at the Egyptian ETRR-2 Research Reactor and has capabilities to produce the following radionuclides:

a) Chromium-51
b) Iridium-192, therapy and industrial gammagraphy sealed sources
c) Molibdenum-99 from fission
d) Molybdenum-99/Technetium-99m generators loading
e) Iodine-131 (solution and capsules) from fission

The facility is as well provided with clean areas and laminar flow for the preparation and loading of Molybdenum-99/Technetium-99m generators and two multipurpose cells for the labelling of compounds and/or the implementation of new processes.

The building of approximately 1200 m2 has conventional, supervised and controlled areas as well as the following fully equipped rooms:

a) laboratories for the preparation and chemical, radio-chemical and biological quality control of products

b) generators loading
c) decontamination
d) wastes storage
e) dispensing and packaging
f) health physics and radio-protection
g) stock and store
h) laundry

The production area comprises eleven shielded cells with the following components/general services:

a) Airtight boxes
b) Shielding
c) Shielded glass viewer or windows
d) Master-slave manipulators or teletongs
e) Process equipment
f) Ventilation
g) Transfer ports and shielded transport containers

Auxiliary equipment such as a calibration hot cell, four shielded glove boxes and three radiochemical hoods with its respective services and vessels for waste collection, were also provided for production and quality control tasks.

Fig. 11 – View of the hot cells front and one laboratory for measurements -RPF Egypt

Fig. 11 – View of the decontamination & preparation cells and the hot cell in the laboratory -RPF Egypt

Fig. 12 – View of the hot corridor -RPF Egypt

Fig. 13 – Tc Generator Calibration and Release -RPF Egypt

9. Irradiation Facilities at ETRR-2 Reactor

As part of the same contract INVAP has supplied AEA of Egypt with the irradiation facilities at ETRR-2 reactor to irradiate targets for the RPF production.

These supplies have comprised design, engineering, manufacturing and procurement, installation and commissioning of these irradiation facilities for both Molybdenum and non-Molybdenum targets.

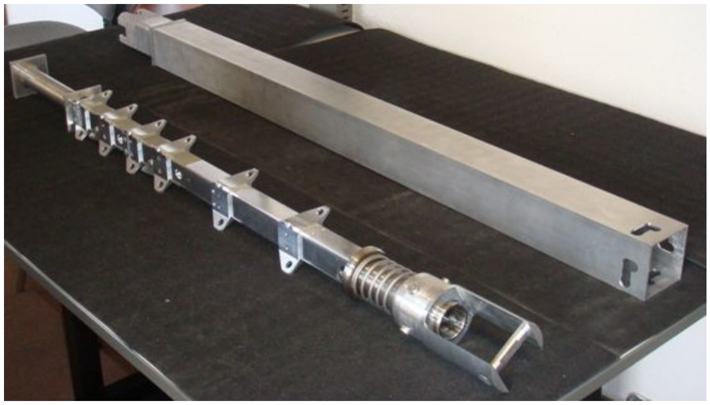

Fig. 14 – Irradiation Rig for the ETRR2 -RPF Egypt

10. Situation Today - RPF

For the past few years the lack of continuous funding and some other AEA internal issues prevented the progress of the project at normal speed. During those setbacks INVAP always maintained the facility and continued with the activities sometimes, just forced to adjust the project pace. In fact cold commissioning activities of the facility were completed long time ago.

Despite the recent political changes in Egypt and the associated uncertainties, AEA and INVAP made a strong effort in moving ahead with the hot commissioning of the RPF which implied the operation of the ETRR2 for sustained periods of time during the week. This has implied a major load on the Egyptian personal trained by INVAP and also in the Argentinean personnel moved temporarily to Egypt (5 to 7 people) under present country situation.

As for today, the irradiation of targets in three different irradiation runs of different duration, have surpassed contractual values: the activity of the molybdenum batches obtained was higher than the nominal value by 5 to 10%. The quality of the product has been higher than the technical requirements and at least equal to the product obtained by CNEA in Argentina.

The other hot runs for the production of Iodine 125, Iridium 192 for industrial sources, Iridium 192 wires for braquitherapy and Iodine 131 have also proved to be successful and beyond the contracted values.

11. Mo-99 Target and Process Project (MOLY) (Australian Nuclear Science and Technology Organization)

After winning the contract for OPAL, INVAP was engaged by ANSTO (Australian Nuclear Science and Technology Organisation) for the supply on a turnkey mode with the design, construction, installation and commissioning of:

a) Irradiation Rigs
b) Mini Plates for the production of radioisotopes
c) the Mo-99 production process
d) A System for waste handling inside cells.
e) Container for filter transport

ANSTO had experience in producing radioisotopes from slightly enriched uranium (2% U235) and therefore had an installation to be reutilized. This imposed a new demand on INVAP effort since existing hot cells already contaminated had to be evaluated and a whole new approach for installing a new process in existing concrete hot cells had to be developed.

Since the hot cells were contaminated by years of production the installation time had to be minimized. Therefore the design, construction and factory acceptance tests of the new process equipment was performed in Bariloche (INVAP´s hometown) and the whole pre - assembled system was sent to Australia for installation. The whole of the design and construction of the process equipment plus its factory acceptance testing was accomplished in 1 and ½ half years. The actual installation work in the hot cells was accomplished in 1 month.

The process for the production of molybdenum is essentially a slightly modified version of the LEU CNEA one which includes the alkaline dissolution of Uranium aluminure targets.

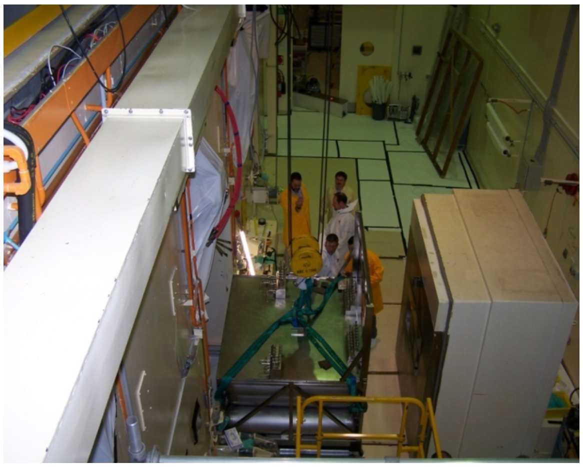

Fig. 15 – Refurbished hot cells with the new process – ANSTO Moly

Fig. 16 – Irradiation Rig for the OPAL - ANSTO

Fig. 17 – New process equipment and Filter Transfer Flask –ANSTO Moly

12. OPAL Moly Project Results

Since its early initial commissioning and runs we understand that today the facility runs 4 batches per week (2 more batches than the original foreseen scheme). Unlike CNEA product which is commercialized and distributed in Argentina and surrounding countries as per Argentinean health care requirements, the product being obtained by ANSTO has been certified by the health care regulators of at least Australia, USA, Canada and Japan. The quality of the LEU product is therefore well established. ANSTO therefore has established as a regular producer and an international player of LEU Molybdenum 99.

This, for INVAP as a designer and supplier, not only praises the quality of ANSTO operations, but also highlights the level of our products, process and engineering.

13. Medical Isotope Production System (MIPS) R&D Program

While the idea of obtaining of radioisotopes from an homogeneous water solution reactor as a source for molybdenum was known for a long time, it had a major issue: to demonstrate that the radioisotopes could be separated from such an active solution with a reasonable efficiency, within a reasonable time frame and with a practical process. It is claimed that a lot of experience and data is available from this type of reactors, but the separation of radioisotopes demanded a full research and development program by itself. With that in mind and Babcock & Wilcox and INVAP started the approach back in 2006 and 2007 and a contract was signed in 2008.

The scope of work for INVAP contemplated the development of a production process to obtain Mo-99 by irradiation of an acid solution in an homogeneous reactor. To the surprise of many, after putting in place a carefully devised program, INVAP accepted a pure R&D contract on a fixed price basis where the RA6 research reactor in Bariloche belonging to Argentina CNEA was the main tool.

The program was staged in phases all aimed at reducing the R&D risks in a such a way that if successful in a stage we could continue to the next one. Based on some previous unsuccessful trials in the 90s the key resided in finding a suitable material that converted into a workable separation column would provide the basis for the first filtering stage of the active homogenous solution.

Generically, the program stages contemplated:

Fig. 18 – Staged Program for B&W R&D of the separation process

The most daring and challenging part of the project included the design, construction, installation, commissioning and operation of a mini loop where a capsule containing the solution is irradiated in the core of the research reactor RA6 in Bariloche, in conditions totally comparable to an homogeneous reactor (irradiation and processing unit) for the development of the project.

For these experiments the RA6 reactor operated in the 2 MW thermal power range for sustained periods of time (5 to 6 days).

Equally important for the accomplishment of the experiments was the design, construction and commissioning of a processing hot cell in the basement of the RA6 building.

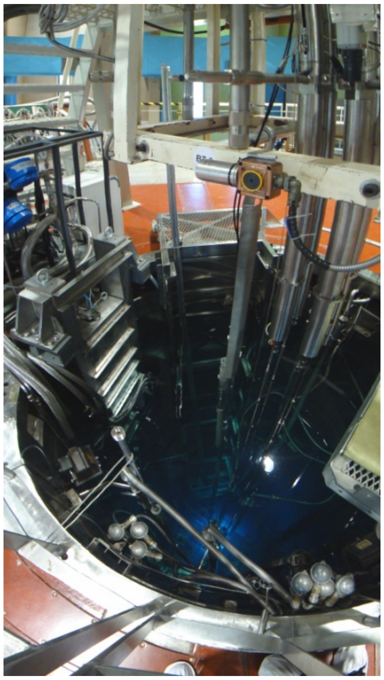

Fig. 19 – RA6 reactor irradiating the solution in the mini loop –B&W project

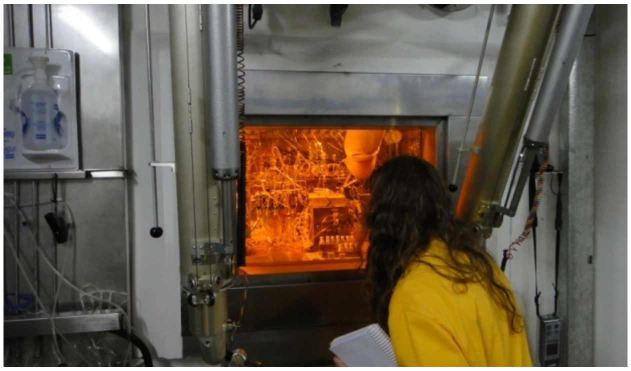 

Fig. 20 – New Hot Cell at the RA6

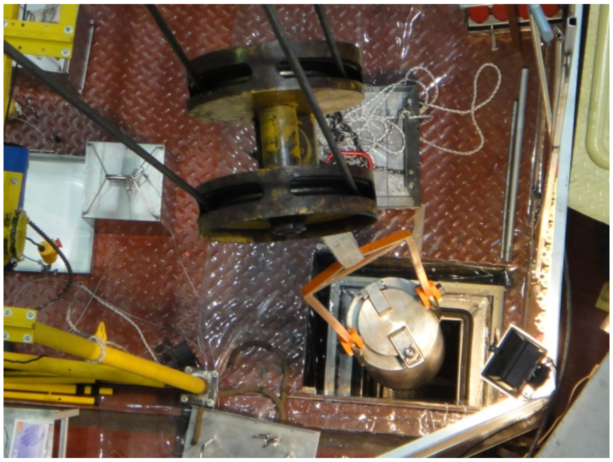

Fig. 21 – Lowering of the Transfer Cask inside the Hot Cell with the Irradiated Solution – RA6

14. MIPS R&D Program Results

The miniloop has been in operation for the past 6 to 8 months and several irradiation and processing runs have been performed (more than 8) where in a repetitive way the molybdenum 99 has been separated from the solution obtaining the specification required.

In a successful way the product was shipped to the US and loaded into a generator where Tc-99 was obtained again within specification.

Therefore and beyond any doubt a process for the separation of molybdenum from an homogeneous solution has been demonstrated and the program can be considered successful.

The R&D program still has some steps to fulfill, all related to improvements and optimization, especially in the efficiency, the manufacturing of the column and the systematization of the process.

It is worth to mention that since inception to initial operation of the miniloop only 2 years elapsed, timeframe which also includes the regulatory submissions and approvals by the Argentinean Regulator (ARN). ARN attitude towards the project was remarkable being able to work in a very collaborative environment with INVAP and CNEA.

15. Other Projects

It is worth to mention some other initiatives and projects INVAP has worked for or is still involved but at a more initial stage.

In the recent past (2008-2009), INVAP provided a study which ended up with the conceptual design of a 3000 6 day Ci plant for MURR (Missouri University Research Reactor).
In more recent times INVAP has been actively working with Coquí Radiopharmaceuticals in a project that contemplates the design, construction and commissioning a research reactor and the associated radioisotope production plant to satisfy a big portion of the needs of the American market or the world. Again INVAP prepared the conceptual approach and an initial proposal for this project.

Much more recently, INVAP started the relationship with IRE from Belgium to assist in the conversion of their production line of radioisotopes from HEU to LEU.

And just a few days ago, CNEA and INVAP signed the Contract were INVAP will start the design of the RA10 research reactor. A much needed facility to replace the venerable RA3, mainstay of the production of radioisotopes in Argentina and soon to be 50 years old.

16. Conclusions

Argentina and INVAP have been, are and will be strong supporters of all the efforts to reduce and eliminate proliferation and nuclear security risks as it is portrayed by all the efforts deployed in 30 years.

INVAP, with every export accomplished (either a research reactor and/or the associated processing facility) all based on LEU has not only demonstrated the technology associated but also has helped avoid its Customers to consider HEU options as an alternative. With the Argentinean domestic production and the projects above mentioned we believe that the LEU based technology for the production of radioisotopes by fission is mature, for any imaginable commercial size.

We agree with the fact that the molybdenum 99 market is suffering a crisis. We also believe that the crisis is an opportunities to have a better future, in this case for human health.

If we revisit Fig. 9 above we will also notice that the Company capabilities and expertise focus on the provision of solutions and not in the production of the radioisotopes. INVAP could be considered as a very unique player in the world of radioisotopes since it devotes its efforts to help the producers deliver with customized LEU based solutions. Not least is the fact that INVAP technology, research reactors and processing plants are cost effective.

Finally, we would like to remark the fact that the projects above mentioned have always been attacked with an integral approach: development, design & engineering, construction, commissioning plus integral regulatory support, all in the same package. This is again a unique capability inside INVAP that minimizes risks for project success.

INVAP wants finally to recognize the support of the Atomic Energy Commission (CNEA) in Argentina, the Argentinean Regulator ARN and the Clients with whom we have always maintained excellent relationships and working environments for the success of the different projects we have been and we expect to be involved.