babcock & wilcox technical services group

Mo-99 Topical Meeting
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INVAP/B&W
Mini-Loop Production of Mo-99

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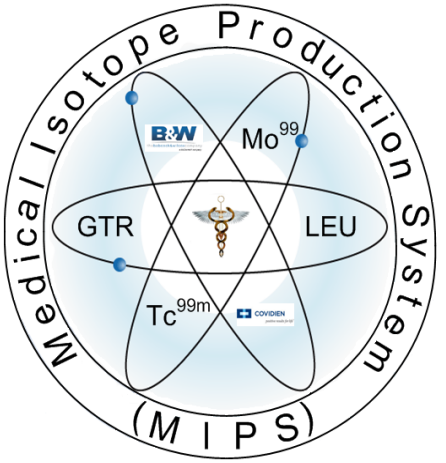
INTRODUCTION

- B&W-TSG is pursuing the development of a Medical Isotope Production System (MIPS) to:

➢ Produce 50% of the domestic need of Mo-99, the precursor to the radiopharmaceutical technetium-99m (Tc-99m).

➢ Support the nuclear non-proliferation mission of the DOE-NNSA Global Threat Reduction Initiative and produce life-saving medical isotopes, such as Mo-99, without using highly enriched uranium (HEU).



Ref: NNSA fact sheet, January 25, 2010

INTRODUCTION

- The US consumes about half of the worldwide production of technetium (Tc)

Ref: Global Nuclear Medicine Market (Medical isotopes / Radiopharmaceuticals) in PET/SPECT Imaging and Therapy – Competitive Landscape, Current trends and forecasts (2010 – 2015), Marketsandmarkets, 2011

- The goal of the MIPS facility is to safely and economically produce 4400 6-day high specific activity curies/week of in-spec Mo-99 through the fissioning of U-235 using a solution of low enriched uranium (LEU) as uranyl nitrate (UN) in an aqueous homogeneous reactor (AHR).

INTRODUCTION

- The MIPS and its use of an AHR are ideal for the purpose of producing life-saving medical isotopes:
  - ➢ LEU is utilized and, thus, supports GTR
  - ➢ The spent fuel is drastically reduced – the U is re-used
  - AHR technology is not new; however, its dedicated and continual use as a production reactor for Mo-99 is new and will require:
  - ➢ Thorough testing, or Proof-of-Principle (P-o-P)
  - ➢ Licensing from the US NRC and product certification from the US FDA

Proof-of-Principle

To provide the P-o-P and provide processing information for the efforts of the NRC and the FDA, B&W-TSG contracted with INVAP to develop, design, construct, and test a “Mini–Loop” concept –

produce Mo-99 by fission of U-235 in solution of
low enriched uranyl nitrate, in a reactor with
similar conditions as the intended industrial facility.

And, in so doing, provide a demonstration of the proposed irradiation technique.

Goals

- Provide insight into the operation and parameters of reactor operations for the production of Mo-99 from the fissioning of U-235
- Assess the effect of radiation on the separation of Mo using a selected inorganic sorbent
- Understand the fate of radioactive and stable species generated from the process in order to promote fuel reuse and develop composition profiles through extraction and into and through purification

Goals (Continued)

- Provide supporting information related to the presence and/or absence of precipitates potentially formed during or after irradiation
- Work toward optimization of the extraction process
- Develop a tailored process for purification
- Perform demonstrations that produce purified Mo acceptable as a precursor to the radiopharmaceutical Tc-99m

EXPERIMENT

- As designed, built, and operated by INVAP
- About 500mL UN solution, 19.75% U-235, is contained in an irradiation capsule, shown below.
- The capsule with solution is placed in the RA-6 reactor pool and irradiated

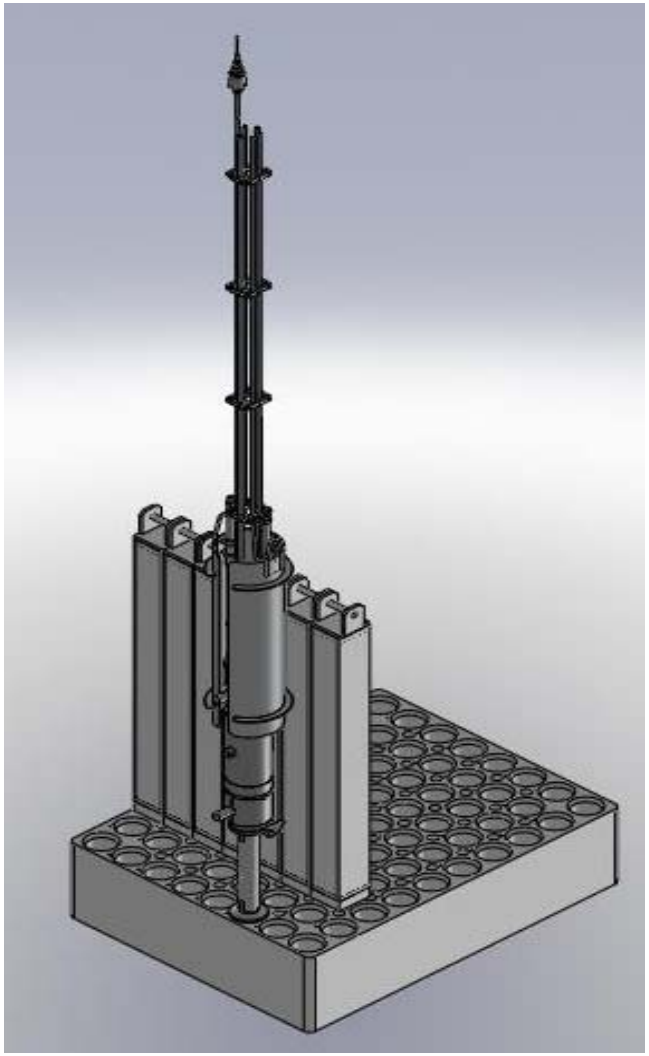
- 1E+12 neutron/cm^2-sec
- 1 kW/L power density
- ~ 1.3 W/cm^2 heat flux (referenced to the wall of the capsule)

EXPERIMENT (Continued)

- Installation into the reactor

EXPERIMENT (Continued)

- Capsule in the grid



EXPERIMENT (Continued)

- The irradiation facility is equipped with:

➢ A system for the treatment of gases

❖ Iodine

❖ Noble gases

❖ Hydrogen

❖ Oxygen

Thus, it has a functional off-gas handling system that at the same time constitutes the prototype for the MIPS.

EXPERIMENT (Continued)

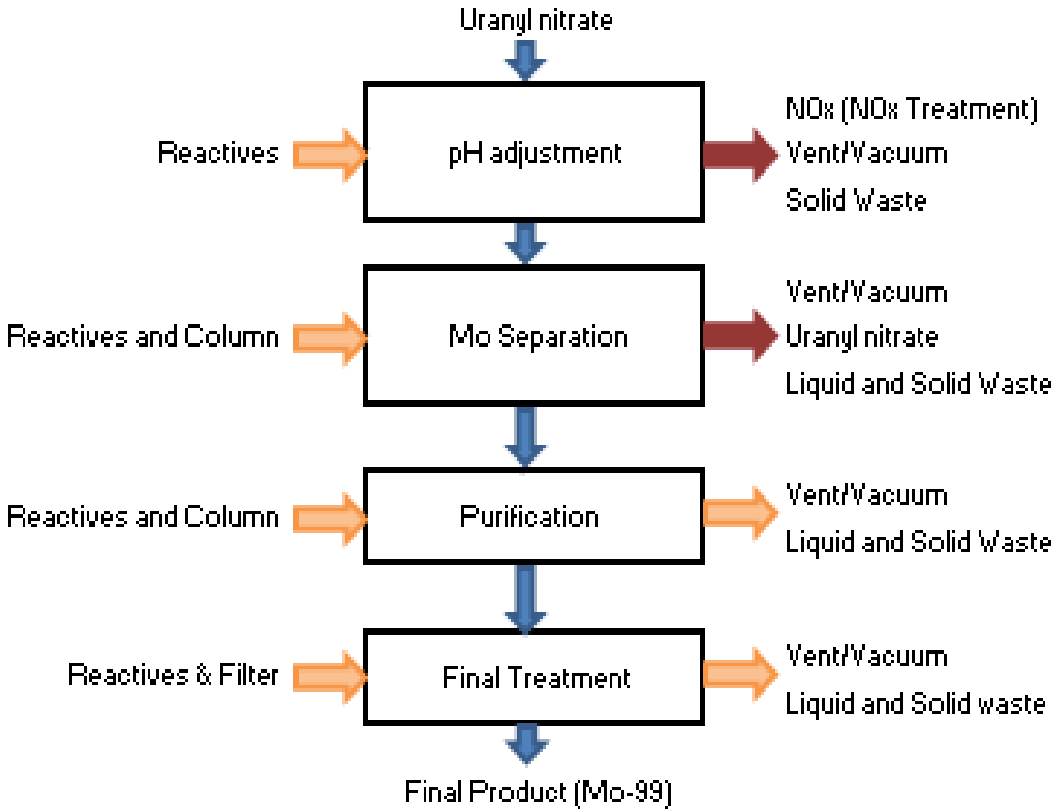
- The Mini-Loop also has:
  - an acid pump for the addition of nitric acid (HNO_3) for the adjustment of pH during irradiation
  - a self-powered neutron detector (SPND) to measure the flux at the exterior surface of the UN solution irradiation capsule
  - a sensor to measure hydrogen concentration in the off-gas

During the irradiations a number of items are measured as a function of time, such as pressure and temperature in various tanks, hydrogen concentration, and temperature at different locations.

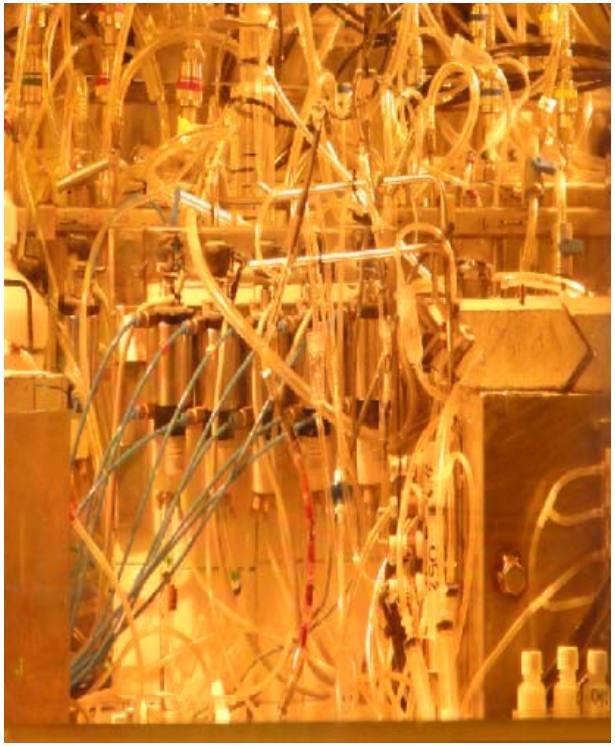
14 sampling points – pH, U, and alpha, beta, and gamma

EXPERIMENT (Continued)

- After irradiation, the UN is transferred via vacuum under the reactor pool water to a transport container, then to a shielded cell for processing as shown in the adjacent flow diagram.



EXPERIMENT (Continued)



All functions in the flow diagram and measurements occur in a single shielded cell.

EXPERIMENT (Continued)

- The partnership of B&W and INVAP helped equip the RA-6 facility. The equipment includes a split glove box with shielding in one side, a gamma spectrometer, an ionization chamber, a UV-Vis spectrophotometer, a liquid scintillation counter, and other basic laboratory equipment.

- INVAP designed the processing system which includes the setup for remote operation of valves, heaters, pumps, and other equipment, all of which are controlled through the use of a programmable logic controller (PLC).

RESULTS

- Six irradiation runs have been performed
- Run #1, #2, and #3 were successful
  - Cross contamination midway through purification caused Run #1 to be out of spec for Zr-95 and Ru-103
- Future runs did not show this contamination issue
- Run #4 was successful through extraction; however, purification was halted due to an unexpected problem in the process line
- Run #5 was successful through irradiation; however, a leak in the transfer line in the reactor pool forced this run to be aborted
- Run #6 was successful with sufficient product produced to load onto a Tc-99m generator, milked, and evaluated

RESULTS (Continued)

These experiments went well, despite one rather large inconvenience – Puyehue – the Chilean volcano!

And, INVAP did a great job pursuing the runs when possible.

RESULTS (Continued)

Streets of San Carlos de Bariloche on June 7th

San Carlos de Bariloche airport on June 7th

RESULTS (Continued)

Attribute/Run	1	2	3	4	5	6
Initial pH	0.5	0.9 - 1.1	0.9 - 1.1	0.9 - 1.1	0.9 - 1.1	0.7-0.9
Irradiation Time (hour)	20	17:50	18	17:05	18	50
HNO_3 added (mL)	0	4	3.3	6.4	2.1	8
Mo-99 produced	Curies	Curies	Curies	Curies	NM	Curies
pH after irradiation	2.5 - 3	0.9 - 1.1	1.1 - 1.4	0.5 - 0.7	--	0.9-1.1
pH before extraction	0.9 - 1.1	0.9 - 1.1	0.7 - 0.9	0.5 - 0.7	--	0.9-1.1
I-131, Ru-103, Te-132, Zr-95, Mo-99	Determined by gamma spectroscopy to follow the process, including sorptive and release behavior					
Mo-99 after extrac (Ci)	Curies	Curies	Curies	Curies	--	Curies
Extrac Efficiency (Mo %)	42	87	87	99	--	97

NM = Not Measured

RESULTS (Continued)

Attribute			Spec[1] ₸	Run #1	Run #2	Run #3	Run #6*
Radiopurity	Gamma	I-131	< 0.05	ND In Spec	ND; In Spec	Detected; In Spec	Detected; In Spec
		Ru-103	< 0.05	Contam Out of Spec	Detected; In Spec	Detected; In Spec	Detected; In Spec
		Te-132	< 0.05	ND In Spec	ND; In Spec	ND; In Spec	ND; In Spec
		Other than Mo-99	< 0.05	NS	NS	Detected; In Spec	Detected; In Spec
		Zr-95	part of Other	Detected; Out of Spec	ND; In Spec	ND; In Spec	ND; In Spec
	Beta	Sr-89	< 6E-04	ND In Spec	ND; In Spec	Detected; In Spec	NM
		Sr-90	< 1.5E-5	NM –Sr-89 is In Spec	NM –Sr-89 is In Spec	NM –Sr-89 is In Spec	NM
	Alpha	Total	< 1E-7	NM	ND; In Spec	ND; In Spec	NM

ND = Not Detected

NS = Not Searched

NM = Not Measured

Contam = Cross contamination of final product.

[1] units are microCi/milliCi Mo-99

* The Mo-99 was loaded onto Tc-99 generator.

The initial tests show that the product meets the required pharmaceutical specification.

₸ *Ind. Eng. Chem. Res.*, 2000, *39* (9), pp 3146–3150

CONCLUSIONS

- The Mini-Loop experiment is a success.

✓ Provided B&W with P-o-P

❖ Demonstrating that high specific activity, pharmaceutically acceptable Mo-99, as a precursor for Tc-99m, can be produced by the fissioning of U-235 in solution

❖ The Mo-99 can be suitably extracted with acceptable efficiency

❖ The initial tests of the Tc-99m off of a generator meets the required pharmaceutical specification

CONCLUSIONS (Continued)

- With its many data collection and measurement points, much is being learned about:

✓ reactor operations

✓ solution chemistry

✓ radionuclide fates

✓ precipitation phenomena

✓ information relevant to the extraction and purification processes

The experimental design is prototypic of MIPS and, thus, its data should be beneficial in the efforts toward licensing by the US NRC and product certification by the US FDA.

ACKNOWLEDGEMENTS

- Oak Ridge National Laboratory for the uranium metal
- Comisión Nacional de Energía Atómica de Argentina (CNEA) facility for converting the uranium metal to U_3O_8
- RA-6 facility for:
  - producing the uranyl nitrate solution
  - operating the reactor and Mini-Loop
- CNEA and the Autoridad Regulatoria Nuclear de Argentina (ARN) to help make this experiment successful.