



Devices to reduce the emission of radioactive noble gases from fission radioisotopes production plants Introduction



History of Fission Radioisotopes Production in Argentina

1967 RA-3 Reactor - HEU fuel elements

- **1977** α , β , γ Hot Cells construction
- **1978 Fission ⁹⁹Mo radiochemical process first experiences**
- **1980 Target development**
- **1985 First commercial production of ⁹⁹Mo HEU targets**
- **1985 Enlargement of the building**
- 1994 New hot cells
- 2000 RA-3 Reactor increases power LEU fuel elements
- 2002 First commercial production of ⁹⁹Mo LEU targets
- 2005 First commercial production of ¹³¹I

Argentina was the first country in the world to change the Molybdenum-99 (⁹⁹ Mo) production process from HEU (high enrichment Uranium) to LEU (low enrichment Uranium), taking into account the Treaty on Non-Proliferation of Nuclear Weapons (NPT) and world initiatives for stricter control of nuclear materials.

Fission ⁹⁹Mo is being produced in Argentina, at the Ezeiza Atomic Center, since 1985. The procedure involved the irradiation of HEU targets with an Uranium-Aluminum alloy "meat" cladded with aluminum.

In the late 90's the Atomic Energy National Commission of Argentina (CNEA) started the development of LEU targets for its ⁹⁹Mo production, in order to replace the HEU targets. In 2002, CNEA started to commercialize ⁹⁹Mo from LEU targets and in 2005 lodine-131 (¹³¹I).

The Medical Isotope Production (MIP) by

Fission release radioactive noble gases into the environment.

These emissions increase the environmental background and could hinder the mission of the **International Monitoring System (IMS)** to detect early a nuclear explosion.

The dissolution of plates with aluminium in a basic medium generates hydrogen. The air is a consequence of the vacuum that must be made in the tanks to move the fluids in the purification stages. The circuits used for H and Air are different as their contents cannot be combined because an explosive mixture could be generated.

Hydrogen is produced by AI during targets dissolution. AI + NaOH + H2O \rightarrow NaAIO2 + 3/2H2



Air	Hydrogen
80%	20%

Relative ¹³³Xe activity between Air and Hydrogen

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Devices to reduce the emission of radioactive noble gases from fission radioisotopes production plants Objectives

Air and Hydrogen

The objective is to show different alternatives to reduce the emission of noble radioactive gases into the environment from a Medical Isotopes Production (MIP) by Fission.

Some devices are applicable to Hydrogen and others to Air and Hydrogen.

A comparative analysis between them is carried out.

Hydrogen

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Devices to reduce the emission of radioactive noble gases from fission radioisotopes production plants Retention Methods for Air and Hydrogen

Evacuated Tanks

Evacuated tanks provide a large volume of storage. More storage represents more decay time and less radioactive noble gases emission to the environment. The gases are stored in tanks, previously evacuated, so that they decay as long as possible.

- Need vacuum pumps
- very little maintenance
- large space

Activated Charcoal

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They are beds of activated carbon that dynamically adsorb Xenon. **Need** change the charcoal periodically and dehumidification of the gas flow. Its performance (DF) improves with:

- lower humidity
- lower gas flow
- larger volume of coal
- lower gas temperature

Devices to reduce the emission of radioactive noble gases from fission radioisotopes production plants Retention Methods for Hydrogen

1.Adsorption

3 Ionisation 4 Diffusion

5. Precipitation 6. Recombination

7 Desorption

2 Dissociation

HYDROGEN CAPTURING DEVICES

The characteristic of hydrogen capturing device is their ability to absorb hydrogen reversibly. When these materials are in contact with hydrogen at a given pressure and temperature a reaction of phase change is produced.

A new compound is formed.

 $M + X/2 H2 \leftrightarrow MHx$

M=hydride forming materials, MHx=formed hydride

- It is a reaction with heat exchange: exothermic when the hydride is formed and endothermic when releases hydrogen
- Need: Heating
- Cooling
- Pump
- Small space

POROUS DEVICES

The hydrogen permeates through the membrane and the impurities (noble gases, nitrogen, etc.) are returned to the tank or are sent to another. The operating conditions need high temperatures to increase hydrogen permeability. It requires differential pressure on both sides of the membrane. - Need: Heating

- Compressor
- Pump
- Small space

COPPER OXIDE REGENERATION: Converts hydrogen to water

Temperature and Pressure of operating: 450 °C,101KPA Need time for regeneration

The converter is more than 95% efficient in reducing the volume of H

- Need: Heating
- Cooling
- Shielding.

Manufactured and installed by Invap Argentina in Radioisotopes Production Plants of Ansto (Australia), Insha (Egypt) and ⁹⁹Mo Production Facility (Mumbay-India).

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Decontamination Factor

Fission MIP producing 400 six days Ci of ⁹⁹Mo, manipulates at Plant 1 10¹⁴ Bq of ¹³³Xe. The Decontamination Factor (DF) is the relationship between this activity of ¹³³Xe and the release of ¹³³Xe into the environment. More storage time of ¹³³Xe in the plant implies a higher DF. Depending on their characteristics and applications, the DF of different retention methods can vary from tens to thousands. (half-life ($t_{1/2}$) ¹³³Xe : 5,247 days)

DF: Initial Activity of ¹³³Xe / Release Activity of ¹³³Xe

weeks	DF
3	16
4	40
5	100
6	256
7	640
8	1600
9	4100
10	8200

DF value based on the weeks that the ¹³³Xe remains in the installation.

Methods	Dimensions	DF
Evacuated tanks (Air – H)	Volume: 6 m ³	40
	Volume: 9 m ³	250
Active Charcoal (Air – H)	Volume: 1m ³ Flow: 0.15 m ³ /h	300
Copper Oxide Regeneration (H)	Volume: 0.2 m ³	250
Porous Devices (H)	Volume: 0.1 m ³	>1000
Hydrogen Capturing Devices (H)	Volume: 0.1 m ³	>1000

The table shows the volumes needed to contain the Xe with the different methods and an approximate value of DF for a production of 400 six days Ci of ⁹⁹Mo

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METHODS	ADVANTAGES	DISADVANTAGES
EVACUATED TANKS	Tested in production Simple, Few devices (vacuum pump) Air and Hydrogen Little Maintenance	Large volume of tanks Ned big space Big shield
ACTIVE CHARCOAL	Tested in production Air and Hydrogen Less volume and higher DF than Evacuated Tanks	Cooling and dehumidification Periodical change of charcoal Need Shield
COPPER OXIDE REGENERATION	Tested in production Low Volume Higher DF	Only Hydrogen. Heating and cooling Need Shield
HYDROGEN CAPTURING DEVICES	Low Volume Highest DF Little shield	Only Hydrogen. Heating ,cooling and compressor Developing-Tested whit ¹³³ Xe
POROUS DEVICES	Low Volume Highest DF Little shield	Only Hydrogen. Heating. cooling and pump Developing-Tested with tracers

Comparative table with some advantages and disadvantages of the different retention methods. The methods can be complementary. For example, use a Copper Oxide Regeneration for Hydrogen and Evacuated tanks or Active Charcoal for Air.

The designer of a Fission MIP should analyse the optimal option according to their needs and possibilities

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Thank you very much

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