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Technical Note

Validation of a New Design of Tellurium Dioxide-irradiated Target

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ABSTRACT

Production of iodine-131 by neutron activation of tellurium in tellurium dioxide (TeO_2) material requires a target that meets the safety requirements. In a radiopharmaceutical production unit, a new lid for a can was designed, which permits tight sealing of the target by using tungsten inert gas welding. The leakage rate of all prepared targets was assessed using a helium mass spectrometer. The accepted leakage rate is $\leq 10^{-4}$ mbr.L/s, according to the approved safety report related to iodine-131 production in the TRIGA Mark II research reactor (TRIGA: Training, Research, Isotopes, General Atomics). To confirm the resistance of the new design to the irradiation conditions in the TRIGA Mark II research reactor's central thimble, a study of heat effect on the sealed targets for 7 hours in an oven was conducted and the leakage rates were evaluated. The results show that the tightness of the targets is ensured up to 600°C with the appearance of deformations on lids beyond 450°C. The study of heat transfer through the target was conducted by adopting a one-dimensional approximation, under consideration of the three transfer modes—convection, conduction, and radiation. The quantities of heat generated by gamma and neutron heating were calculated by a validated computational model for the neutronic simulation of the TRIGA Mark II research reactor using the Monte Carlo N-Particle transport code. Using the heat transfer equations according to the three modes of heat transfer, the thermal study of I-131 production by irradiation of the target in the central thimble showed that the temperatures of materials do not exceed the corresponding melting points. To validate this new design, several targets have been irradiated in the central thimble according to a preplanned irradiation program, going from 4 hours of irradiation at a

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power level of 0.5 MW up to 35 hours (7 h/d for 5 days a week) at 1.5 MW. The results show that the irradiated targets are tight because no iodine-131 was released in the atmosphere of the reactor building and in the reactor cooling water of the primary circuit.

1. Introduction

Two major sources of artificial radioisotopes are accelerators and reactors. Radioisotopes produced in research reactors represent a large percentage of the total radioisotopes needed. A reactor offers a large volume for irradiation, simultaneous irradiation of several samples, economy of production, and the ability to produce a wide variety of radioisotopes.

Accelerators are generally used to produce those isotopes that cannot be produced by reactors or that have unique properties [1].

Iodine-131 has been used as a radioactive drug in nuclear medicine departments for decades. This radiopharmaceutical is necessary for functional exploration of adrenal glands, kidneys, and bladder cancer cases; whole-body scan; metabolic irradiation of metastases; and treatment of hyperthyroidism. There are two methods of I-131 production in nuclear research reactors. One involves the irradiation of tellurium targets and the other uses uranium targets.

Production of iodine-131 from tellurium dioxide, TeO_2, requires a target that meets the safety requirements (homogeneity of welds, leak tightness, resistance to irradiation conditions, etc.).

The purpose of this study is to validate a new design of TeO_2 target that will be used for the production of iodine-131 in the central thimble irradiation position of the TRIGA Mark II research reactor at Centre National de l'Energie des Sciences et des Techniques Nucléaires, Rabat, Morocco.

2. Production of iodine-131

2.1. Production of ^{131}I by (n, γ) reaction using ^{130}Te

Iodine-131 is produced by irradiation of TeO_2 targets in a thermal neutron flux environment according to the (n, γ) nuclear reaction, as shown in Fig. 1 [1].

As natural tellurium contains 33.8% ^{130}Te nuclide, there is no special need to use an enriched tellurium target except in nuclear research reactors with a low neutron flux or when high specific activity is required. However, natural tellurium contains other isotopes that generate unwanted radionuclides, causing difficulties during processing and storage [2].

2.2. Failures of TeO_2 irradiations

Several incidents of TeO_2 cans failures during irradiation have been observed. A severe incident occurred at the BR2 reactor during the irradiation of TeO_2 in an aluminum can sealed by cold welding. It was caused by the failure of a TeO_2 can, which led to the release of iodine-131 in the reactor building. This incident was explained by the reduction of TeO_2 by aluminum at a high temperature, as shown by the following reaction:

$$3TeO_2 + 4\ Al \rightarrow 2Al_2O_3 + 3Te \text{ (energy released: 582 kJ/mol)} \quad (1)$$

It seems that the reaction starts slightly before the melting point of aluminum and from 700°C it becomes extremely bright and highly exothermic. The molten aluminum disks placed on either side of TeO_2 pellets reduce the oxide, generating high heat (3,500 cal per disc). This resulted in a high heat flux to the surface of the capsule, estimated to be at least 130 W/cm^2. Critical flux burnout was then reached, which explains the TeO_2 target failure [3]. An incident also occurred in the TRIGA research reactor at Energy Research Establishment, Dhaka, Bangladesh, during an irradiation process in the dry central thimble. Very high radioactivity was detected. After physical investigation, a melted Pyrex ampoule containing TeO_2 powder was found within the damaged aluminum can that contained the Pyrex ampoule. The main cause of failure of the Pyrex ampoule was identified as the presence of ^{10}B isotope in the ampoule, which captured neutrons and generated more heat. After that incident, the reactor was operated regularly by loading TeO_2 in the wet central thimble at 2 MW power instead of 3 MW by using quartz in place of Pyrex ampoules due to its high melting point [4].

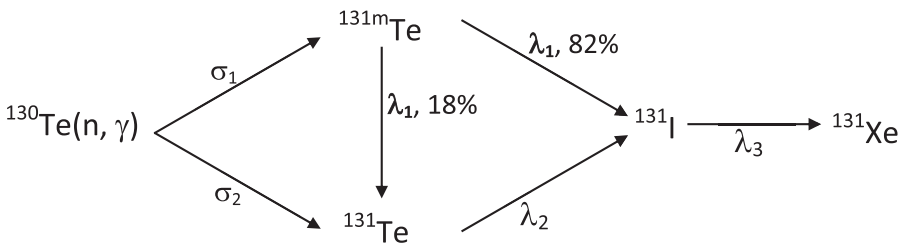

Fig. 1 – I-131 production process by activation of Te-130 isotope.

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