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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].

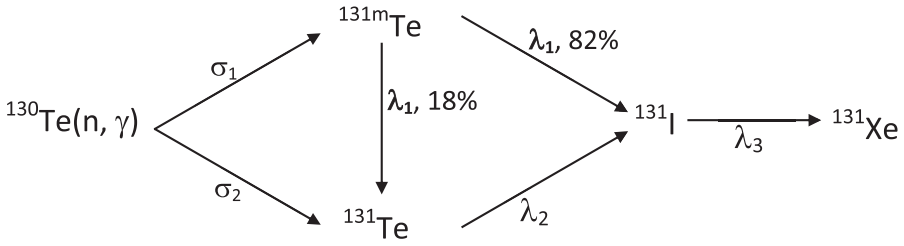

Fig. 1 – I-131 production process by activation of Te-130 isotope.

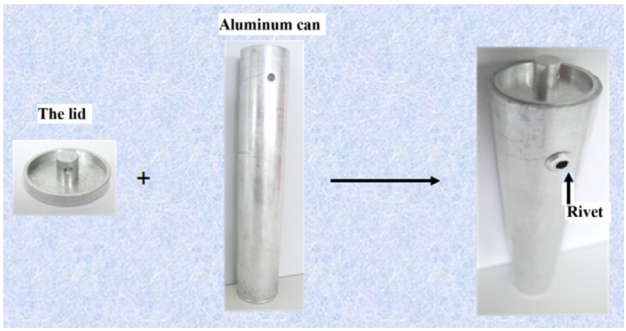

Fig. 2 – Old design of the target.

2.3. *Design of the new target*

The available target shows leaks around the rivet used for sealing the target by cold welding, as illustrated in Fig. 2.

To meet the safety requirements, a new lid for the can was designed. It involves changing the location of perforation of the can in the old design to another location that will not cause any problem during the final phase of tungsten inert gas (TIG) welding. The possible solution to the problem of leaks encountered in the old design using the rivet for sealing is to change the design of the lid, thus avoiding the use of the rivet that does not ensure tight sealing of the target. The lid should be provided with a chimney that can evacuate expanded air when pressure increases within the target during TIG welding, as shown in Fig. 3.

For safety, 15% of the total space in the quartz ampoule was kept for air, to contain the gas produced during the irradiation process. Fig. 4 gives an overview of the irradiated target.

Table 1 lists the mass of TeO_2 material and dimensions of irradiated target, and Table 2 lists the proprieties and thermophysical parameters of materials.

2.4. *Heat transfer analysis*

The study of heat transfer through the target was conducted by adopting a one-dimensional approximation, under consideration of the three modes of transfer—convection, conduction, and radiation. The quantities of heat generated by

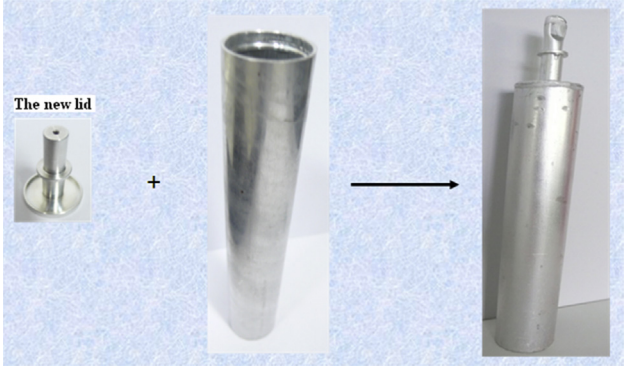

Fig. 3 – New design of the target.

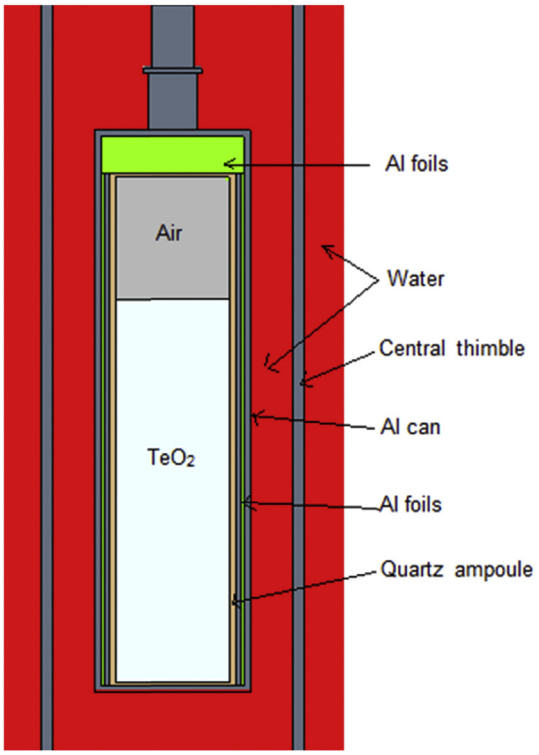

Fig. 4 – Overview of the irradiated capsules.

Table 1 – Dimensions of TeO_2-irradiated target.

TeO_2 weight (g)	Internal radius of quartz (cm)	External radius of quartz (cm)	Internal radius of Al can (cm)	External radius of Al can (cm)
65	0.8	0.9	1.05	1.20

Table 2 – Proprieties and thermophysical parameters of materials.

Material	Density (g/cc)	Thermal conductivity (W/m/K)	Emissivity (ε)	Melting point (°C)
Water	0.992	0.67	–	–
TeO_2	5.67	1.1	0.89	733
Aluminum	2.7	230	0.18	630
Quartz	2.65	4	0.89	1710

Table 3 – Neutron and gamma heating calculated by MCNP transport code.

Heat (W/g)	Neutron	Gamma	Total
TeO_2	0.8329	0.3475	1.1804
Quartz	0.0311	0.2760	0.3071
Aluminum (can + foil)	0.0145	0.2720	0.2865

MCNP, Monte Carlo N-Particle.

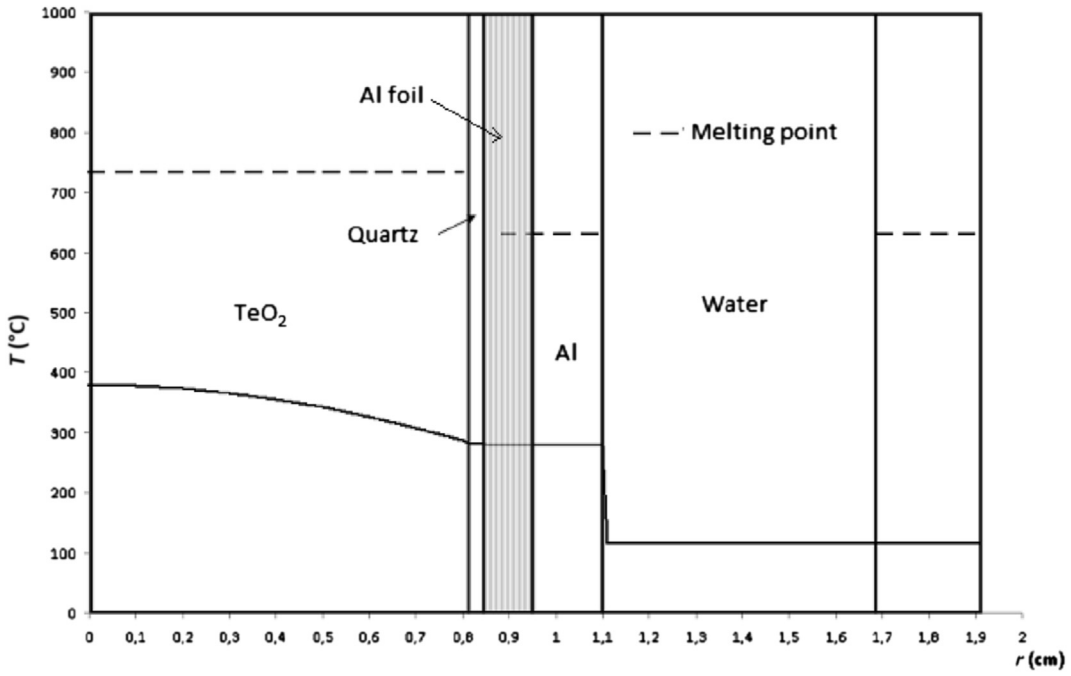

Fig. 5 – Temperature distribution within the irradiated capsules.

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. The results of the neutron and gamma heating are summarized in Table 3.

Using the heat transfer equations according to the three modes of heat transfer, the thermal study of iodine-131 production by irradiation of the target in the central thimble showed that the temperatures of materials do not exceed their corresponding melting points, as illustrated in Fig. 5 [5]. The temperature in the middle of TeO_2 reaches 378°C.

The design proposed in this study will avoid such incidents as stated earlier. TeO_2 will be sealed in a quartz ampoule as a first barrier to avoid an exothermic reaction between Al and TeO_2 at temperatures above 660°C, the melting point of aluminum. The filled ampoule will be wrapped with an aluminum foil to avoid the air gap between the quartz ampoule and the aluminum can, in order to facilitate heat transfer and avoid any mechanical risk that may lead to rupture of the quartz ampoule. The wrapped quartz ampoule will be placed in an aluminum can and then sealed by TIG, as shown in Fig. 3.

3. Materials and methods

The target consists of high-purity TeO_2 powder (99.4%) purchased from Sigma Aldrich, St. Louis, MO, USA. Pure quartz ampoules were purchased from Isotope Technologies Dresden GmbH, Dresden, Germany. Aluminum cans and lids (purity: Al 99.5 type: EN-AW 1050 A) were purchased from Hydro Aluminum Rolled Products, Hamburg, Germany. The aluminum foil is of high purity (>97.5%).

3.1. *Target preparation*

3.1.1. *Cleaning of aluminum items*

(1) Set the irradiation cans and their lids into a scouring basket with cover.
(2) Close the scouring basket in order to prevent the irradiation cans from swimming on the surface of the scouring bath due to the generation of hydrogen during scouring.
(3) Dip the scouring basket with irradiation cans and lids into a hot (70–90°C) aluminum scouring bath (consisting of ~10% sodium hydroxide solution with a suitable additive, to prevent precipitation of hard solid deposits in the scouring tank). The hotter the scouring bath, the shorter the scouring time. The scouring process roughens, defects the surface, and removes the surface oxide layer from aluminum cans.
(4) Dip the scouring basket two to four times into cold tap water to rinse the scoured irradiation cans.
(5) To neutralize the scoured irradiation cans, dip the scouring basket into half-concentrated nitric acid (7M nitric acid) at room temperature for ~10 seconds. Undesired alloy components, such as copper, magnesium, silicon, etc., are eliminated.
(6) Finally, dip the scouring basket into hot tap water (at ~80–90°C) and let the irradiation cans dry in air [6].

3.1.2. *Purification of TeO_2*

TeO_2 of high purity can be used as the target material without pre-purification, but purification is indispensable due to interfering impurities such as tellurium nitrate and other

compounds that are volatile at temperatures up to 800°C. These impurities can affect the quality of sodium iodide solution to be produced. Purification of TeO_2 needs to be carried out as follows [6]:

(1) Wash a mass of TeO_2 into a glass-filter funnel using bidistilled water at least until the used wash water shows a neutral pH.
(2) Dry the TeO_2 in a covered glass-filter funnel overnight.
(3) Transfer the moderately dried TeO_2 into porcelain crucibles, dry it carefully at 100–150°C.
(4) Melt it for 1 hour to evaporate impurities that are volatile below the melting point of TeO_2.
(5) After melting, cool down and crush the TeO_2, which often shows a green or yellow-brown color.
(6) Grind the TeO_2 lumps in a pebble mill.
(7) Sieve the powder and collect the 0.15–0.2 mm fraction.
(8) Store the sieved powder in a desiccator.
(9) Test the powdered sample for constant weight after heating at 150°C.

3.1.3. *Filling quartz ampoule*

Fill each quartz ampoule with 65 g TeO_2 powder.

3.1.4. *Sealing quartz ampoule*

Seal the filled quartz ampoule by the oxyacetylene welding process.

3.1.5. *Tightness quality control of the sealed quartz ampoule*

The bubbles leak test is a quick way to check the tightness control of the quartz ampoules, which is performed as follows [7]:

(1) Completely immerse the welded ampoule in liquid nitrogen for a period of 5 minutes.
(2) Transfer the ampoule into the test liquid (normally ethanol).
(3) The ampoule is observed over a period of at least 1 minute.
(4) The ampoule is tight if no bubbles emanate from it.
(5) Wrap the controlled quartz ampoule with aluminum foil and insert it into the aluminum can.

3.1.6. *Sealing the aluminum can*

Welding consists of joining two metal pieces by establishing a metallurgical atom-to-atom bond, which is different from a joint held together by friction or mechanical interlocking. This metallurgical atom-to-atom bond is achieved by the application of heat and sometimes pressure [8].

In this context, an aluminum lid has been designed with a long chimney, as illustrated in Fig. 3. The first purpose of the chimney is to permit TIG welding of the lid to the can without deterioration of the welding bath. Therefore, during the TIG welding, the air inside the can is expanded by heat and comes out through the chimney. Second, the long chimney permits sealing of the top part of the lid by pressing (cold welding), which permits TIG welding of the top part of the lid without deterioration of the welding bath. The purpose of cold welding of the top part of the lid before TIG welding is to maintain the expanded air inside the target during TIG welding and protect the welding bath. Sealing of the aluminum can is performed as follows:

(1) Wrap the controlled quartz ampoule with an aluminum foil and insert it into the aluminum can.
(2) Weld the aluminum lid to the can by a TIG welding machine.
(3) The weld of the target is controlled for leaks by compressed air at 4 bars in a water bath.
(4) Cold weld the extremity of the lid by a hydraulic press at 2 bars.
(5) Weld the extremity of the lid by TIG welding.

3.1.7. *X-ray examination of welds*

A film is placed behind the sealed target and exposed to an X-ray beam. Defects in welds are shown by a black color.

3.1.8. *Tightness quality control of sealed can*

Tightness of the aluminum can is tested by two methods.

(1) *The bubble leak test:* it is a quick control of the tightness of the aluminum can. The control is performed as follows: (i) completely immerse the welded can in liquid nitrogen for a period of 5 minutes; (ii) transfer the can to the test liquid (normally ethanol); (iii) observe the can over a period of at least 1 minute; and (iv) the can is tight if no bubbles emanate from it.
(2) *Leak test by helium mass spectrometer*. This control is performed using a helium leak detector (HLT 560) purchased from Pfeiffer Vacuum, Berliner Str. 43, 35614, Asslar, Germany, as follows: (i) the sealed target is placed in the charging chamber. The charging chamber is purged by a vacuum pump and then pressurized with helium to a pressure of 5 bars for 30 minutes; (ii) the charging chamber is then depressurized and the target is transferred to the test chamber; (iii) helium is discharged through a helium mass spectrometer detector to quantify the leakage rate; and (iv) the acceptance criterion is a leakage rate of lower than 10^{-4} mbr.L/s. This criterion was recommended during the International Atomic Energy Agency (IAEA) mission in the frame of the TC Project RAF 4022, conducted at the Moroccan TRIGA Mark II research reactor to advise on safety aspects related to the production of iodine-131, from 28 December 2011 to 29 December 2011 in Rabat, Morocco.

3.2. *Heat effect on sealed target*

The studied targets are heated at different temperatures between 100°C and 600°C for 7 hours in an oven, carbolite BWF 12/13 type.

3.3. *Irradiation*

The target is irradiated in the central thimble of the TRIGA Mark II research reactor, where the measured thermal neutron flux at 2 MW power is $\Phi_{th} = 7.0110^{13}$ n/cm^2/s.

3.4. Iodine-131 extraction

The irradiated target will be opened in a special hot cell. The quartz ampoule is transferred in a special steel beaker and cut off by the manipulators. The irradiated target material for iodine production will be filled into porcelain crucibles and set into the furnace in which the internal surface is a heated quartz tube. Then [^{131}I] iodine is separated from radioactive TeO_2 by a dry process (volatilization) at ~695°C and transferred into the iodine traps by carrier air. The carrier air containing gaseous [^{131}I] iodine passes through an electrically heated separator (a quartz tube with quartz filling) set at 200°C, which reliably retains TeO_2 impurities and dust particles [6].

The purified gaseous iodine-131 is then absorbed into a weakly alkaline buffer solution (0.019M $NaHCO_3$ and 0.0082M Na_2CO_3) in the iodine traps (a 0.01M NaOH solution is also recommended for use as an absorber solution). The obtained product is weakly alkaline [^{131}I] NaI stock solution. The recovery efficiency is evaluated to be 80–90%. The iodine separation apparatus was purchased from Isotope Technologies Dresden.

3.5. Radioactivity measurement

The radioactivity of alkaline [^{131}I] NaI stock solution is measured in a dose calibrator, VIK-202, purchased from Veenstra Instruments, Madame Curieweg 1, P.O. Box 115, Joure, 8500 AC, Netherlands.

4. Results and discussion

4.1. Bubble test in liquid nitrogen

All prepared targets were bubble controlled in liquid nitrogen. The results showed no bubbles emanating from the targets.

4.2. Leak test by helium mass spectrometer

The leak rate of targets was evaluated by a helium mass spectrometer. The results showed that the leak rate for all targets was below 10^{-6} mbr.L/s, which complies with the criteria of acceptance.

The new design of the target provides a better seal with a leak rate within the specified range (leakage rate $\leq 10^{-4}$ mbr.L/s). All the prepared targets (100%) were tight.

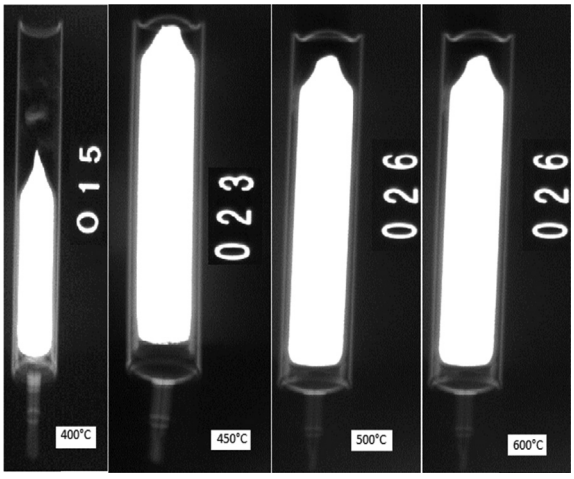

Fig. 6 – X-ray examination of the targets after heat treatment.

4.3. Heat effect on the tightness of the target

This study was conducted with five sealed targets that were heated in an oven at temperatures between 100°C and 600°C for 7 hours. The leakage rate is measured after cooling. The results showed that the leak rate for all targets was below 10^{-6} mbr.L/s, which complies with the criteria of acceptance.

4.4. Visual inspection of targets

Heating the target from 100°C up to 400°C has no effect on its shape, as shown by target 015 in Fig. 6. However, heating at 450°C, 500°C, and 600°C caused swelling at the top and bottom covers of the target, as shown by targets 023 and 026 in Fig. 6.

4.5. Bubble test in liquid nitrogen

After heat study, all targets were bubble test controlled in liquid nitrogen.

The results showed that heating targets up to 600°C for 7 hours did not affect their tightness (no bubbles released), despite the appearance of swelling at the top and bottom covers at temperatures of 450°C, 500°C, and 600°C.

4.6. Leakage rate test by helium mass spectrometer

The helium leakage rate of the studied targets was evaluated by a helium mass spectrometer. The leakage rate at different temperatures, even at 600°C, is acceptable since it remains significantly lower than the acceptance limit (10^{-4} mbr.L/s).

4.7. Radiographic examination

X-ray examination was performed on three targets (015, 023, and 026) after heat treatment at 400°C, 450°C, and 500–600°C, respectively, as shown in Fig. 6.

Radiographs showed good homogeneity in welds with low penetration.

Table 4 – Validation assays of the new design of TeO_2 target.

Assay no.	Weight of TeO_2 (g)	Power (MW)	Irradiation time (hr)	Cooling time (hr)
1	30	0.5	4	48
2	30	1	7	68
3	65	1	2.5	48
4	65	1	3	68
5	65	1	7	48
6	65	1.5	35	68

4.8. *Validation of the new design by irradiation in the central thimble*

To validate this new design, several targets have been irradiated in the central thimble at power levels ranging from 0.5 MW up to 1.5 MW. Table 4 lists all assays that were conducted to validate this new design of the TeO_2 target in the Moroccan TRIGA Mark II research reactor.

The irradiated targets were tight, no deformations were observed on irradiated targets, and no iodine-131 was released in the atmosphere of the reactor building or in the reactor cooling water of the primary circuit.

Monitoring of iodine at the top of the reactor tank is controlled by the iodine monitor FHT 1702 L with a detection limit of < 1 Bq/m^3 at 100 nSv/h and < 40 Bq/m^3 at 100 μSv/h [9].

The release of iodine into cooling water of the primary circuit was controlled by taking samples before and at the end of the irradiation cycle from the cooling water of the primary circuit. The samples were analyzed by a gamma spectrometer that belongs to the safety and security pole. The results show that no iodine was detected in the cooling water of the primary circuit.

5. Conclusion

Tightness of the target with the new design has been demonstrated in an oven at 600°C despite the appearance of deformations above 450°C. Tightness of the target with the new design has been demonstrated at irradiation conditions in TRIGA Mark II reactor's central thimble at a power level of 1.5 MW. No iodine-131 was released during irradiations, and no deformations were observed in irradiated targets. The low penetration of the welds may be overcome by reducing the clearance between the cover and the capsule.

Conflicts of interest

The authors have no conflicts of interest.

Acknowledgments

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