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Analysis of thermal incident due to sticking of the al-can containing irradiated TeO_2 target to the bottom of the dry irradiation channel of the 3MW TRIGA MK-II research reactor

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Abstract

Radioactive Iodine-131 is one of the most important medical radioisotopes (RI) that are being used at fourteen (14) institute of nuclear medicine and allied sciences of the country. It is routinely produced by irradiating TeO_2 powder loaded into a heat sealed quartz vial called "target" in the Dry Central Thimble (Dry CT tube) of the 3 MW_{th} TRIGA MK-II nuclear research reactor of Bangladesh Atomic Energy Commission. The target is kept inside the standard irradiation canister/can made of Aluminum, called "Al-can". A few reportable thermal incidents occurred during the irradiation of the TeO_2 powder at full power due to lack of proper evaluation of safety of in-core irradiations. Of them, sticking of the Al-can containing 40g irradiated TeO_2 target to the bottom of the Dry CT tube at full power (3 MW) operation of the reactor was one of the major thermal incidents in November 2008. The stuck Al-can with the Dry CT tube in the reactor core was lifted up using an innovative handling tool designed and developed by the author. The physical and visual investigations have identified several weaknesses related to the thermal incident. The thermal problem of the incident has been addressed elaborately and then has attempted to solve using experiments as well as numerical analyses. Two cases (Case-1; Dry CT tube and Case-2; Wet CT tube) have been analyzed both numerically and experimentally. It has found from the numerical analyses and experiments that temperature assessment considering 50g TeO_2 power at 3MW full power operation of the reactor over the walls of the quartz vial and Al-can are about 86 and 70^0C, respectively which is far below the melting point of these materials for the case of Wet CT tube. On the other hand, temperature over the walls of the quartz vial and Al-can are about 520 and 370^0C, respectively for the case of Dry CT tube. The shape and size of the quartz vial and the area of its contact point with Al-can plays a very vital role in rising the temperature of the target material. 50g TeO_2 target irradiation at full power operation of the reactor under dry condition of the CT tube may cause vial failure due o excessive temperature rise that cause melting of the TeO_2/Al-can. It is thus recommended to use a Wet CT tube replacing the existing Dry CT tube for irradiation of more target materials for routine production of RIs in the research reactors without repeating the thermal incident.

Keywords: RI production, TeO_2 target, Dry irradiation channel (Dry CT), Safety analysis, Target geometry, Wet irradiation channel (Wet CT)

Introduction

$3MW_{th}$ TRIGA MK-II research reactor of Bangladesh is being used for routine-wise radioisotope production (RI) and other R&D purposes since 1986. Presently the requirement of reactor based radioisotope has increased significantly in the fourteen (14) nuclear medicine centers of the country; as such it is strongly felt that some measures should be taken in this regard. One of the ways to achieve the goal is to increase the amount of TeO_2 from 30 to 50g. On 26 October 2008, 40g of TeO_2 powder, sealed in a quartz vial, was irradiated in the Dry Central Thimble (Dry CT tube) of the reactor at 3MW power level for production of I-131. The location of the Dry CT tube in the reactor core is shown in Fig.1. The physical dimensions of quartz vial, Al-can, Dry CT tube and other operating conditions in and around the stuck Al-can are as follows. The quartz vial was 1.75 mm thick. The inner and outer radii were 9.38 and 11.13 mm, respectively. The height of the vial was 75.2 mm. The vial was kept in an Al-can. The Al-can was inner and outer radii of 12.05 and 12.45 mm, respectively where the height of the Al-can was 95.1 mm. The Al-can was in rest on the top of a stopper plate of aluminum. This plate was 3.81 cm thick. Under the plate, there was water of 60 ^{0}C. The Al-can was kept in the Dry CT tube, which is also made of aluminum (Grade: T6-6061 alloy) as shown in Fig. 2. The inner and outer radii of the Dry CT tube were 16.94 mm and 19.06 mm, respectively. The height of the Dry CT tube was 8 meter. Outside the Dry CT tube, water at about 40^{0}C was circulated to remove the generated heat in and around the irradiating target.

The Dry CT tube had a dog-leg bend at its middle, which prevents direct streaming of radiation from the reactor core. Under this condition, the irradiated Al-can was stuck with the Dry CT tube. For this, the reactor operation was suspended for several months. In addition, a few reportable thermal incidents occurred during the irradiation of the TeO_2 powder at full power due to lack of proper evaluation of thermal assessment [2, 7]. The detailed description of thermal incidents mainly sticking of the Al-can and its removal techniques from the Dry CT tube are given in Section 2.

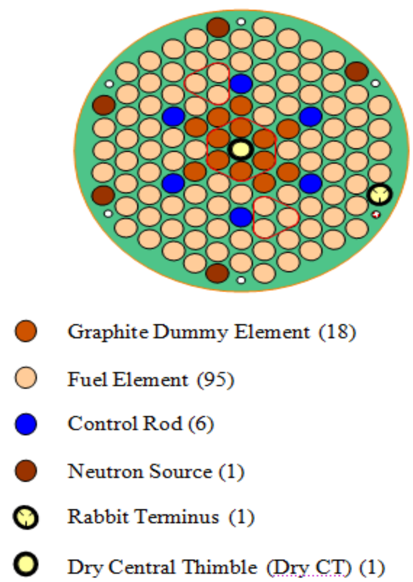


Figure 1 Reactor core

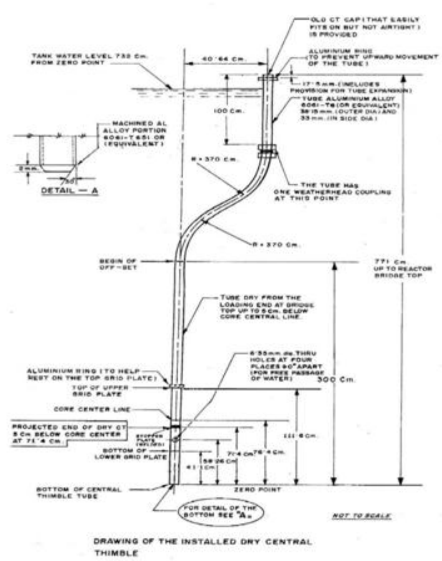


Figure 2 : Schematic drawing of Dry CT tube for I-131 production

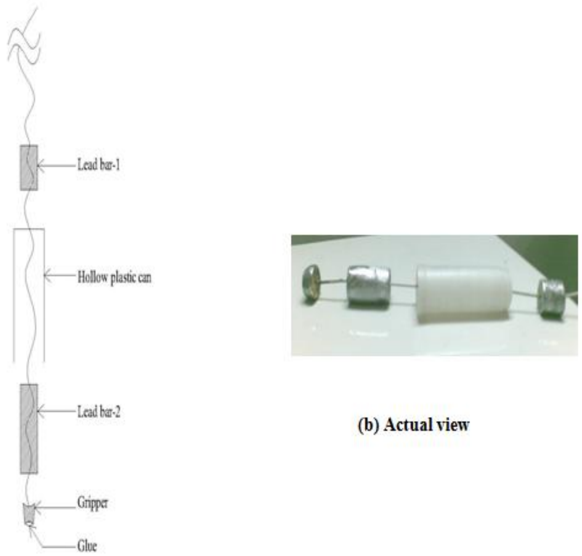
In all these thermal incidents, the heat sealed quartz vial was found to be broken probably because of excessive heat generation and pressurization inside the heat sealed quartz vial [3, 4]. The real causes of these failures are of prime importance to investigate from the safety point of view of the reactor. For this purpose, a basic experimental set up was made and experiments were performed to understand the heat transfer mechanism and also to understand the real situation of thermal incidents [5]. From the experiments, it was measured the gross temperature rise over the heated surface of the quartz vial as well as the Al-can. Few tests results were found to be encouraging. Moreover, numerical studies were also performed to verify with the experimental results [6]. After establishing the methods and techniques for safety analysis of different irradiating target materials in the existing Dry CT tube, possibility of installing a Wet CT for irradiation of TeO_2 powder will also be examined for increasing the RI production program with assured safety. The paper mainly focuses about the techniques on how to lift up the stuck irradiated Al-can to the bottom of the Dry CT tube in the reactor core and also find out the root causes of these thermal incidents under the Two Case Studies though verification of experimental and numerical ones. Case study-1 represents dry condition of the CT tube with holes in the Al-can and Case study-2 represents water filled in the CT tube with holes in the Al-can.

Description of thermal incident

The first thermal incident took place in March 2002 when the reactor was operated for 8 hours at full power to irradiate 50g of TeO_2 powder in the Dry CT tube. The TeO_2 powder was filled in a heat sealed Pyrex vial (in stead of heat sealed quartz vial) and then placed inside an Al-can. The Al-can was then placed in the Dry CT tube. When the irradiated Al-can was removed from the Dry CT tube, it was found that its lower part melted. Further investigation revealed that the lower part of the Pyrex vial was also melted and about 30g (out of 50g) of the irradiated TeO_2 powder was left inside the Dry CT tube. After the incident, the contaminated Dry CT tube was replaced by a Wet CT tube.

After that in 2007 a few minor thermal incidents occurred again during irradiation of TeO_2 powder in a heat sealed quartz vial in the Dry CT tube. In these incidents, the heat sealed quartz vial was broken, color of the TeO_2 powder was got blackish and lower part of the Al-can was deformed.

Subsequent to this, another major thermal incident took place on 2 November 2008. A heat sealed quartz vial containing about 40g of TeO_2 was irradiated in the Dry CT tube for about 28 hours in 5 days (26-30 October 2008) at a power level of 3 MW. At the end of irradiation and after a natural cooling period of about 54 hours (during the weekends) reactor operators attempted to lift the irradiated Al-can from the Dry CT tube using GA (General Atomics, USA) supplied specimen handling tool on 2 November 2008. At that time the cap (upper part) of the Al-can came out. But the body of the Al-can with irradiated quartz vial containing 40g of TeO_2 powder remained inside the Dry CT tube. Several tools (electromechanical gripper, pneumatic gripper, etc.) were used to take the sticking irradiated Al-can out of the Dry CT tube but all the efforts were unsuccessful. Finally, on 13 January 2009, it was possible to take the Al-can out along with the quartz vial containing irradiated 40g TeO_2 powder using an innovative handling tool as shown in Fig. 3 designed and developed by the author [1]. Figure 3(a) shows the schematic diagram of the innovative handling tool whereas Fig.3(b) shows the actual view of it. The tool used some adhesive (with an appropriate applicator) for gripping the Al-can. Adhesive was kept in the curved cavity of the gripper as shown in Fig. 4 that was made by a steel nut. The gripper was welded to a long threaded-steel-rope. Two lead-bars 1 and 2 of different sizes and weights were placed as shown in Fig.3 for applying necessary impact force on the gripper such that it (the gripper) sits well on the sticking Al-can. The hollow plastic can work as a guider to guide the gripper during its travel inside the Dry CT tube. The traveling speed of the gripper was controlled manually by gradual releasing the steel-rope.



(b) Actual view

(a) Schematic diagram

Figure 3 Innovative handling tool

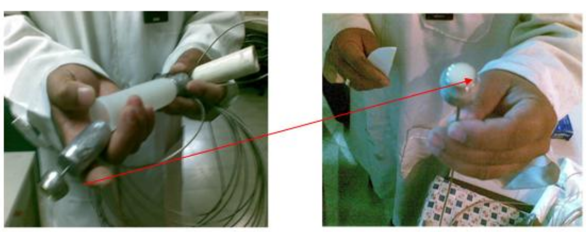


Figure 4 Gripper filled with adhesive

Once the gripper was placed on the Al-can (as well as on the quartz vial), it was left there for about 48 hours so as to allow sufficient time for adhesive to develop strong bondage with the Al-can and the quartz vial. After that the gripper along with the Al-can and quartz vial was lifted up applying strong pulling force through the threaded-steel-rope.

Investigation of the unusal event

To review and analyze the causes of the vial failure incident of 2002, an International Atomic Energy Agency (IAEA) expert mission was conducted at research reactor facility in 2003 [8]. The expert identified several causes of the incident by analyzing numerical calculations using 1-dimensional GENTC code. The main cause of the failure of the vial, as identified by the expert was the presence of B-10 in the Pyrex. It was unusual to use Pyrex vial instead of quartz vial for any irradiation purpose in the reactor core. That analysis was not enough to attack the complicated three dimensional (3-D) problem.

Figure 5 Lifted Al-can with some white materials sticking to its bottom

Again to review and analyze the causes of the another major vial failure incident of 2008, as the Al-can came out with the help of the innovative but very simple, appropriate, and effective handling tool (cost around 10 USD), it was seen through physical and visual investigations that a scale like sintered material, white in color, caused the Al-can to remain stuck to the Dry CT tube bottom plug. This can be seen in Fig. 5. Investigations have also revealed that the upper part of the Al-can was marked with black stains while the lower part was covered with a scale like white colored material, which is thought to be coming from the TeO_2 powder that deposited at the bottom of the Dry CT tube from previous incidents that resulted failure of quartz vials containing TeO_2 powder. The powder formed a layer between the Al-can and the bottom stopper plug of the Dry CT tube and offered significant resistance to the flow of heat. With the rise of temperature, the powder was melted and made the Al-can sticking to the bottom of the Dry CT tube. During irradiation at 3MW, nuclear heating (neutron + gamma) gradually heated up the target, which did not have good heat dissipation path connected to the reactor cooling water. Most of the heat generated in the TeO_2 target first flows through the TeO_2 itself, then through the bottom part of the quartz vial, then through the bottom of the Al-can and after that through the bottom stopper plug of the Dry CT tube, which is a piece of Al-plate having a thickness of about 3.81cm.

The incident did not make any damage to the reactor core or cause any significant release of radioactivity to the environment. The surface gamma dose rate of the removed Al-can (with the quartz vial containing 40g TeO_2 in it) was about 4.29 mSv/hr. The incident was timely informed to the regulatory authority. Necessary cleaning up operation was carried out jointly by the reactor operation personnel and the health physics group so as to make the Dry CT tube ready again for irradiation of TeO_2 powder for I-131 production.

Observed causes and corrective actions

In all of these thermal incidents, the heat sealed quartz vial was found to be broken probably because of excessive heat generation and pressurization inside the heat sealed quartz vial. Inadequate heat transfer was identified as the main cause of these thermal incidents in the TRIGA Research Reactor. As the amount of heat generated depends on the quantity and quality of TeO_2 target being irradiated, the temperature rise was found to have direct links with these parameters. The geometry of the quartz vial also plays a vital role for transferring the generated heat effectively into the surrounding. Considering the limitations of the Dry CT tube for irradiating TeO_2 targets, it has been decided to convert the Dry CT tube into a Wet CT so as to enhance the production of I-131 with added safety [7]. The author took initiatives to perform rigorous safety analysis study so as to not repeating the thermal incident. The following section describes the detailed of the safety assessment studies of the simulated target.

Safety assessment

Experiments and Numerical Analysis

The safety analysis specially temperature assessment on simulated target (Al-can with quartz vial containing 50g TeO_2 powder) both experimental and numerical was performed. The experimental set-up was built up as shown in Fig. 6.

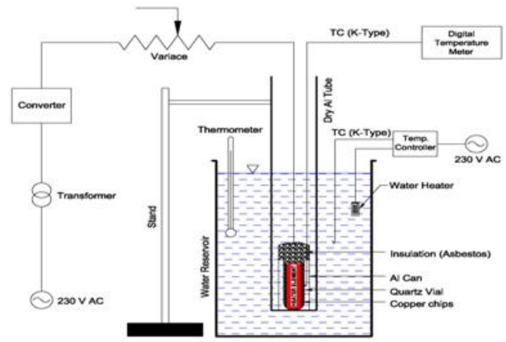


Figure 6 Schematic diagram of experimental set up for Dry/Wet CT tube

The simulated target consists of a DC heating element which can supply heat as equal to the generating heat of 50g TeO_2 powder at 3MW full power operation of the reactor. The simulated target was kept inside the Al-tube that was the same grade of the Dry CT tube. The Al-tube was first kept in dry condition, called "Dry CT" tube and then the tube was filled in water, called "Wet CT" tube. The Al-tube was kept in the water reservoir. A k-type thermocouple was used to measure the surface temperature of the quartz vial. It was also measured the surface temperature of the Al-can using another k-type thermocouple. Water temperature of the reservoir was kept constant at 40 ^{0}C using a temperature controller and its temperature was monitored by using a glass thermometer. A variac was used to control the DC power supply of the heating element. The set-up was used to perform several experiments so as to understand the heat transfer mechanism and to visualize the real situation that led to the incidents. A few experimental snap-shots are shown in Fig. 7.

Figure 7. A few experimental snap-shots

Figure 7(a.1) shows the simulated target consisting of a DC heater that works as heating element. Copper chips were added around the gap of the heating element in order to transfer the generated heat quickly through the quartz vial. Original quartz vial was used to hold the heating element and copper chips. The opened-top surface of the quartz vial was closed with the help of adhesive/asbestos. Then the simulated target was kept inside the perforated Al-can as shown in Fig.7(a.2) and the cap was fitted on to the top of the Al-can with the help of adhesive. A few holes were drilled at the height of 20 mm and 70 mm of the Al-can. It is to be mentioned that original quartz vial and Al-can (regularly used in the reactor for RI production) were used in preparing the simulated target. Experimental condition is shown in Fig.7(b) which shows an Al-tube, water reservoir, temperature controller, a DC power supply system and a digital temperature meter. Experiments were carried out for the two cases that include Dry CT tube and Wet CT tube with perforated Al-can inside the simulated target. Boiling phenomenon as shown in Fig.7(c) was observed when simulated target was kept inside the transparent plastic tube with filled in water instead of Al-tube. The measured temperatures at different locations over the target are given in Table 1.

A numerical study by CFD (Computational Flow Dynamics) was also carried out for the two cases (Case study-1 & Case study-2) so as to simulate the situation of heat transfer that actually occurs in the Dry CT tube. For the Wet CT tube with holes model, the water column height was kept constant at 2m to check whether it has an effect on the centerline temperature. For the case of numerical analysis, the vial was modeled as a heat source with a heat generation rate of 2.46×10^3 W/kg at 50g TeO_2 powder. The outside wall of the CT tube was modeled with a constant temperature of 40 ^{0}C. The bottom surface of the stopper plate was modeled with a constant temperature of 60 ^{0}C. Except these, all the walls were assumed of "no slip" condition. The top of the CT tube was assumed to be at 30 ^{0}C. Figure 8 shows the 3D model with grid mesh of the simulated target with perforated Al-tube.

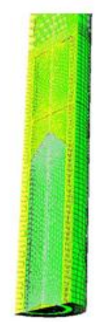


Figure 8. 3D Model geometry with grid mesh

In Fig. 9, for the Dry CT tube with holes model, shows a centerline temperature of 557 ^{0}C by the RNG $k-\in$ model. The centerline temperature drops to the minimum for the Wet CT with holes over the Al-can when water height of 2m inside the Al-tube which is shown in Fig.10. The center line temperature was dropped from 557 to 139 ^{0}C for the RNG $k-\in$ model. The temperature over the walls of the quartz vial and Al-can is also of great interest.

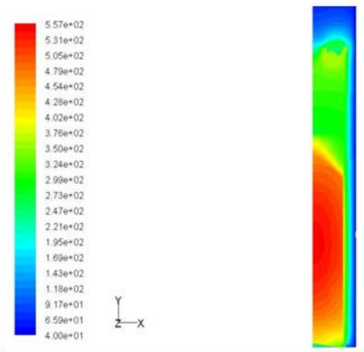


Figure 9 : Contours of static temperature (^{0}C)- Case-1: Dry CT tube with holes over the Al-can and the RNG $k-\in$ model

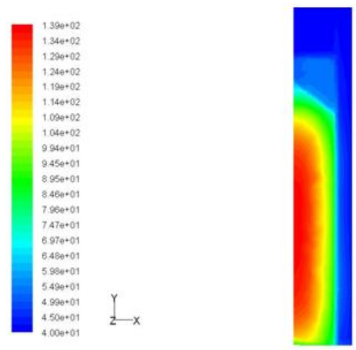


Figure 10 : Contours of static temperature (^{0}C) - Case-2 : Wet CT with holes over the Al-can and the RNG $k-\in$ model (Water Height : 2m)

Table 1: Temperature at different locations over the simulated target (Al-can, quartz vial, TeO_2 powder)

Case study	Centerline temperature of the TeO_2 powder (^{0}C)		Temperature at the outer surface of the quartz vial (^{0}C)		Temperature at the outer surface of the Al-can (^{0}C)	
	Result obtained through CFD modeling	Result obtained from experiments	Result obtained through CFD modeling	Result obtained from experiments	Result obtained through CFD modeling	Result obtained from experiments
Case 1	557	-	510.88	520	366.14	370
Case 2	139	-	85.87	84	66.8	70

It is observed from Table 1 that temperature over the walls of the quartz vial and Al-can are about 86 and 70 ^{0}C, respectively which is far below the melting point of these materials. Temperatures found by CFD analysis and experimental methods at different locations over the simulated target are given in Table1. There are some discrepancies between numerical analyses and experimental results. The discrepancy may be due to measurement error during experiments. Safety of the reactor core may be disrupted if any melting occurs either in the TeO_2 powder/ Al-can in the Dry CT tube when both the quality and quantity of the TeO_2 powder and reactor power do not maintain properly. More experiments are needed to gather more measurement data at different hot-spots of the target and the CT tube.

Conclusion

Primary investigations based on physical and visual have identified several weaknesses related to thermal incidents. It has realized that during irradiation at 3 MW_{th}, nuclear heating (neutron + gamma) gradually heated up the target, which did not have good heat dissipation path connected to the reactor cooling water. As a result it (target with Al-can) was formed a solid mass with the Dry CT tube because of the excessive temperature rise that caused melting of the TeO_2 powder which ultimately forced to break the quartz vial and then melting the Al-can. Rigorous safety analysis, particularly with regard to temperature and pressure rise, is a must before putting any target material inside the in-core terminus of an irradiation facility. As the qualities of commercially available quartz vials and TeO_2 powder may vary from batch to batch, it is difficult to ensure safety of the targets by a single analysis. The shape and size of the quartz vial (larger size) and the area of its contact point with Al-can plays a very vital role in the rise of temperature of the target material loaded into the quartz vial. It has found from the numerical analyses and experiments that temperature over the walls of the quartz vial and Al-can are about 86 and 70 ^{0}C, respectively for the case of Wet CT tube which is far below the melting point of these materials. On the other hand, temperature over the walls of the quartz vial and Al-can are about 520 and 370 ^{0}C, respectively for the case of Dry CT tube. Thus it is recommended that Wet CT tubes are preferable to Dry CT tubes for irradiation of more target materials for routine production of RIs. This is because of excessive temperature rise. The detailed numerical analyses along with some further detailed experiments can really provide a better cooling method for the CT tube of the reactor which consequently can help increasing I-131 production rate. In-core irradiation safety not only ensures the safe operation of the research reactor but also strengthens the RI production program with assured safety.

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