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A NEW ^{124}Xe IRRADIATION SYSTEM FOR ^{123}I ROUTINE PRODUCTION AT THE 30 MeV IPEN-CNEN/SP CYCLOTRON

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ABSTRACT

Since 2001 the Nuclear and Energy Research Institute (IPEN/CNEN-SP, Brazil) has produced about 2.5 mCi/μAh of ^{123}I weekly using a manual irradiation system fully developed by its researchers. Ultrapure ^{123}I has been produced and distributed to hospitals and clinics where several diagnostic imaging procedures are done for thyroid, brain and cardiovascular functions. Due to the short half-life and emission of low-energy photons, this radioisotope becomes suitable for diagnosis in children. Currently IPEN researchers are involved in the development of a new fully automated irradiation system dedicated to ^{123}I routine production employing enriched xenon gas (^{124}Xe) as the target material. This new system consists of a target port, a water and a helium cooling system, a cryogenic system, an electric power system, a control and process monitoring unit composed of a supervisory software connected to a Programmable Logic Controller (PLC) via personal computer. In this new concept, there is no need for human interference during radioisotope production, reducing the possibility of eventual failures or incidents involving radioactive material. In this way, with this new system, a specific yield of approximately 3.5 mCi/μAh per irradiation is expected and this will meet a large part of the national demand for this important radioisotope. In the present work will be presented all the technical and constructive aspects of this new system as well as the results obtained in the irradiation of tests.

1. INTRODUCTION

^{123}I is a radioisotope that has been widely used worldwide for imaging diagnosis of thyroid, brain and cardiovascular system disorders [1-3]. Due to its short half-life and emission of low-energy photons, it is well suited for diagnoses in children [4].

Until the late 1990's The Nuclear and Energy Research Institute (IPEN) had a cyclotron accelerator (CV-28 - *The Cyclotron Corporation*) and started the experimental production of ^{123}I employing the route shown in (1) by irradiating a solid target of enriched tellurium oxide [5,6].

$$^{124}Te\left(p,2n\right)^{123}I \tag{1}$$

In 1998, with the acquisition of a new cyclotron (Cyclone 30 - *Ion Beam Applications*), the ^{123}I routine production was started at IPEN using an irradiation system fully developed by its researchers, employing the route shown in (2) [7-12].

$$^{124}Xe\left(p,2n\right)^{123}Cs \rightarrow {}^{123}Xe \rightarrow {}^{123}I \tag{2}$$

Until 2009 all ^{123}I routine productions were performed using this irradiation system, but because it was a manual system, there was a need for human interference in many phases of the process, increasing the chances of failure and incidents with radioactive material.

For this reason, a new fully automated ^{123}I production system has been developed. This work presents the constructive aspects of the developed system as well as the results obtained with the first irradiations.

2. SYSTEM DESCRIPTION

In order to make the ^{123}I production more safely, minimizing human interference and the risks of incidents with radioactive material, an automated system was developed that allows the ^{124}Xe transfer to the target port, gas recovery after irradiation, removal of ^{123}I produced, and cleaning and drying of the irradiation system.

The new system design consists of: the target port, a closed pressurized helium-cooled circuit for molybdenum windows, a cryogenic system for ^{124}Xe transfers before and after irradiation, a vacuum system to maintain the irradiation system at low pressure, temperature and pressure gauges, an electric power system for the vacuum pumps, valves and heating resistors activation. All such systems are remotely controlled and monitored by a Programmable Logic Controller (PLC) via an RS232/USB interface with a personal computer employing a supervisory software developed in *MS Visual Basic 6*® specifically for that purpose. Figure 1 below shows a block diagram of the developed system.

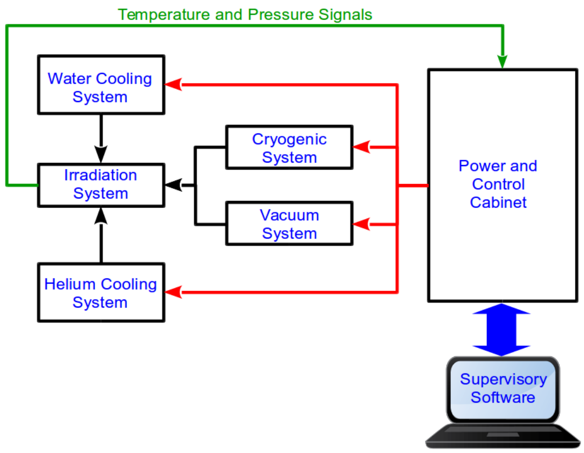

Figure 1: Irradiation system block diagram.

2.1. The Target Port

The new target port consists of a cone-shaped radiating chamber, an external cooling jacket, a beam dump at the bottom of the target port, a front collimator and a set for mounting the molybdenum windows separating the irradiating chamber from the Cyclotron beam line [13,14]. Figure 2 show a sketch of the target port.

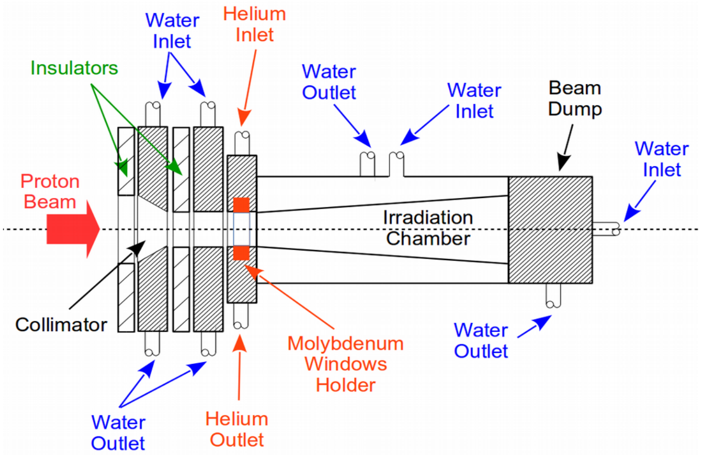

Figure 2: Sketch of the target port for ^{123}I production.

2.2. Water Cooling System

The Figure 3 shows a schematic view of the ^{124}Xe target port cooling system. Cooling is performed independently in three parts of the irradiation system: collimator, target port body and beam dump. The same system also cools the heat exchanger and the helium compressor of the molybdenum window cooling system. The target port has an independent water purge system used only for preventive and/or corrective maintenance purposes.

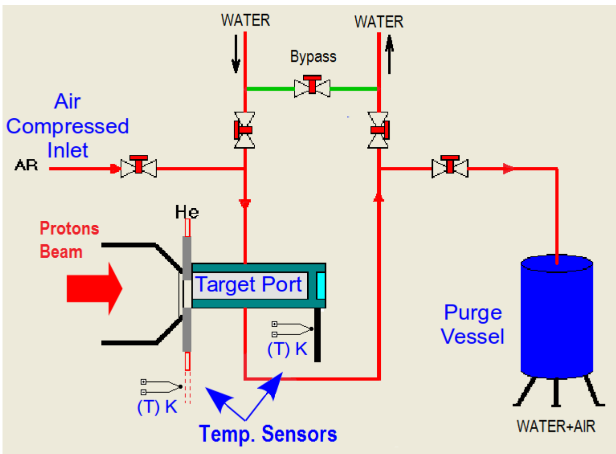

Figure 3: Water cooling system.

2.3. Helium Cooling System

The Figure 4 shows a schematic of the helium cooling system employed to cool the target port molybdenum windows. Initially this system should be purged with gas helium flow about 15 seconds. After purge in this way the system works in loop. After passing through the molybdenum windows the helium gas temperature is monitored by means of a thermocouple installed in the gas outlet pipe.

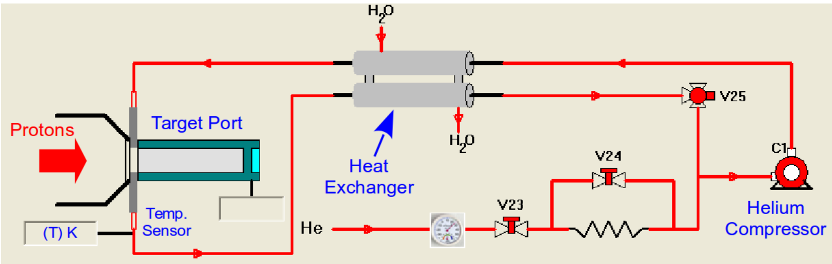

Figure 4: Helium cooling system.

2.4. Cryogenic System

For the ^{124}Xe transfer both into the target port and its future recovery after ^{123}I production, a cryogenic system was developed as shown in Figure 5. Liquid nitrogen can be transferred to either the main cooling vessel or the intermediate cooling vessel near the target port. Inside these cooling vessels are housed the storage ^{124}Xe cylinders and connected to each of these cylinders there is a set of power resistors used for the liquid nitrogen evaporation and also to facilitate the transfer of the gas by thermal gradient. Likewise, around the target port body, there is another set of power resistors used to heat the water which facilitates the ^{123}I adsorption which is adhered to the inner walls of the irradiating chamber. For gas transfer to be efficient, in addition to the temperature gradient, the piping must be under vacuum (around 10^{-3} mbar). In this case the gas is transferred into the target port and the subsequent recovery after irradiation. Vacuum pump B1 maintains the gas loading and unloading tubing under vacuum prior to and after irradiation, and vacuum pump B2 is responsible for drying and maintaining under vacuum the tubing through which water^{+123}I is withdrawn.

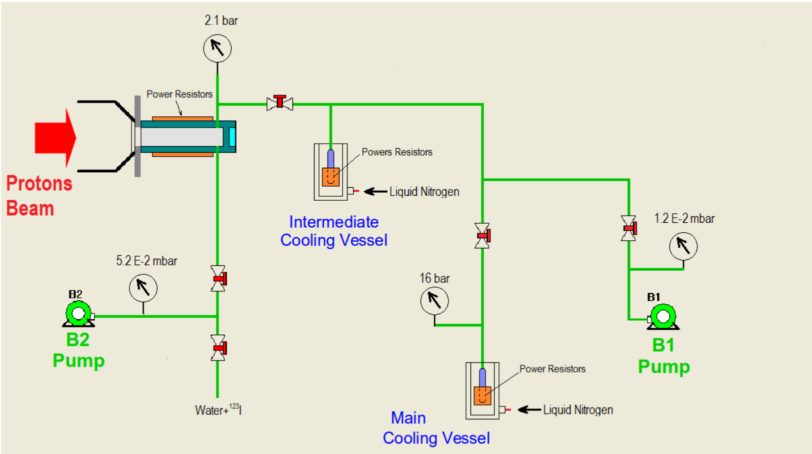

Figure 5: Cryogenic system.

2.5. Power and Control Cabinet

During xenon irradiation, the radiation levels inside the room are very high mainly due to the high energy neutrons emitted during the target bombardment. For this reason, in the Electrical System design it is necessary to consider the effects of this radiation on the electric circuits

and their components, since the radiation can cause degradation and irreversible damages in its structures, compromising the system durability and reliability.
In this way, the Electrical System (distribution, command and monitoring) was divided or subdivided into units considering the protection of its components, either through its installation in remote locations, and even the use of shields, such as used in some sensors. Also, when possible, electromechanical components, thermocouples, etc., which are more resistant to radiation were used.

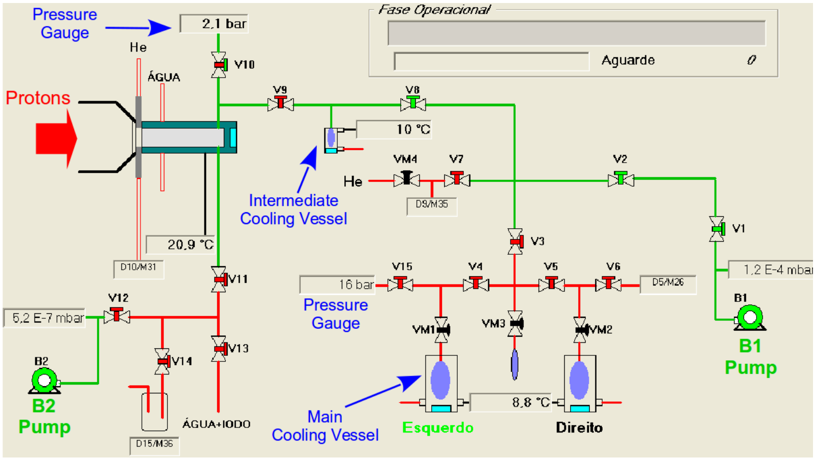

Figure 6: Supervisory software window used for ^{124}Xe transfer and recovery.

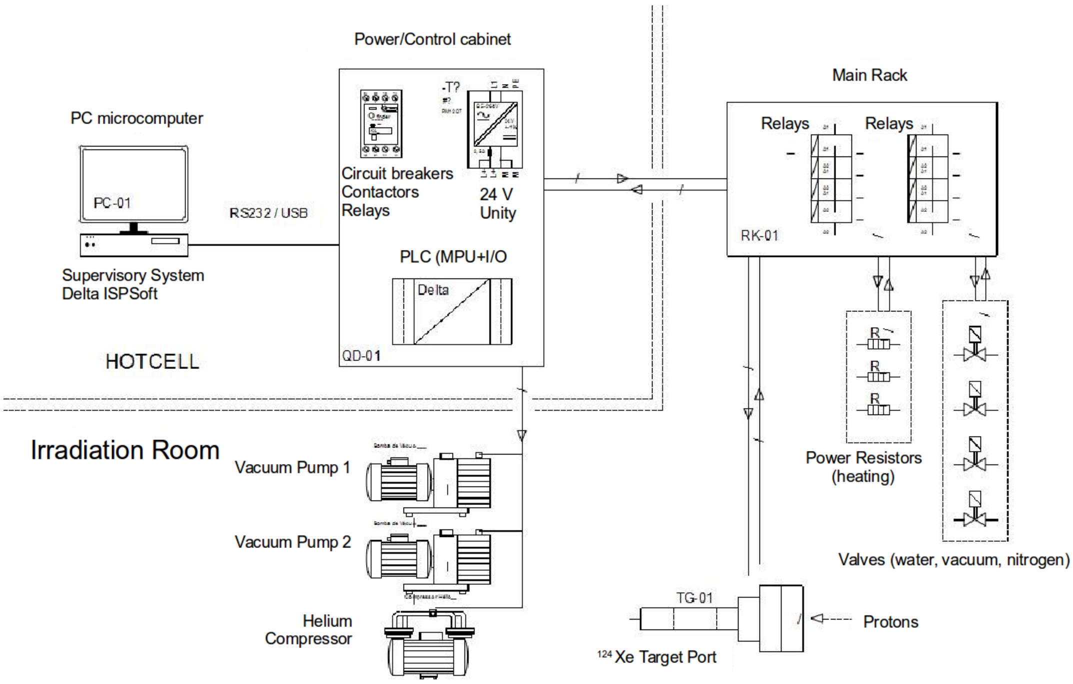

Figure 7: Power system for ^{123}I production.

The Electrical Power System consists of a main rack located at the irradiation room and a control panel interconnected to a microcomputer located in an external room. The main rack, via remote control signals (from the control panel), energizes directly or indirectly (through electromechanical relays) the respective solenoids valves (pneumatic, water, nitrogen, etc.), as well as heating resistors. Also in this rack are connected the vacuum pumps and the helium compressor. As for the control panel, responsible for sending the respective control signal to the Main Rack, it consists of circuit breakers, contactors, thermal relays and the control unit itself, i.e. a Programmable Logic Controller (PLC) composed of a main module and six I/O extension modules (digital inputs and outputs and analog inputs for sensor connection) that perform the acquisition of system process status (positions, temperatures, pressures, etc.), as well as such as the control of various actuators (electric, pneumatic) of the Main Rack. Also, a PC microcomputer, with a supervisory software developed in *MS Visual Basic®*, connected to the PLC via RS-232/USB interface, monitors the process. Figure 7 above shows a summary of the Electrical System developed for this application.

2.6. Process Control Software

The entire ^{123}I production process is remotely controlled by a software supervisory developed in *MS-Visual Basic 6®*. Figure 6 above shows to the system operator the supervisory software main screen where the process control windows appear. According to the access level the system allows the execution of tasks according to two security modes described in Table 1.

Table 1: Safe access levels of ^{124}Xe irradiation system

Safety Mode	Description
Maintenance	All devices unlimited access mode. Used in maintenance activities. Access allowed for qualified personnel only.
Operational	Limited access only to the ^{123}I production process. The operator can access all command buttons related to the process step and monitor the relevant parameters of each step (temperature, pressure, etc.)

In ^{123}I routine productions, the Operational Safety Mode must be used allowing the operator to control the process steps. The next item will be described in more detail the steps involved in the production of ^{123}I using the Automatic System developed at IPEN.

3. PROCEDURES FOR ^{124}Xe IRRADIATION AND ^{123}I PRODUCTION

Table 2 below provides the description of the operational steps necessary for performing a ^{123}I routine production.

Table 2: Operational steps for ^{123}I production

Steps	Description
Target Load	The first step is turned on the B1 and B2 pumps (Figure 6) to remove the helium gas present in the system by preparing the chamber to the ^{124}Xe gas transfer. The ^{124}Xe is cryogenically transferred from the main storage cylinder to the intermediate cylinder and into the target port irradiating chamber.
End of Bombardment (EOB)	At the end of the bombardment, the system is maintained in standby mode (approx. 6 h) until the ^{123}I formed adheres to the inner walls of the target port irradiating chamber.
Target Irradiation	During irradiation, the system is constantly monitored by means of the pressure, temperature and flow gauges present in the system. If any of the parameters are outside of the defined operating limits, the system issues an alert to the cyclotron operator and the irradiation is immediately suspended.
^{124}Xe Recovery	After ^{123}I formation, the ^{124}Xe remaining is cryogenically transferred from the target port irradiation chamber directly to the main storage cylinder.
^{123}I Withdrawal	The irradiation chamber is filled with sterile water and the target power resistors set is turned on to heat the water to approximately 343 K for 20 minutes thereby removing the ^{123}I adhered to the inner walls of the irradiation chamber. Then the chamber is cooled to room temperature and the water plus ^{123}I solution is transferred to the hot cell by pressurizing the irradiation chamber with helium gas.
Target Port Cleaning	For about 2 minutes a continuous flow of helium gas circulates through the irradiation chamber whose function is to remove humidity present after the ^{123}I transfer. Thereafter, the B2 pump (Figure 6) is turned on to complete the cleaning and drying process of both the irradiation chamber and the tubing. And at the same time the B1 pump (Figure 6) is turned on for cleaning the other branch of the tubing.
System Stand-by Mode	After all previous steps the target port irradiating chamber must be filled with 2 bar of helium gas and maintained at this condition until the next irradiation. Then all systems can be turned off.

3.1. Target Load

When starting the target port loading, the water cooling system valves must be opened. The helium cooling system should then be purged for approximately 15 seconds to clean transfer line and compressor and valve tests. The system is commissioned for the next step.

As mentioned above, the helium gas inside the irradiation chamber must be removed prior to the xenon gas loading, this is done by turn on the vacuum pumps B1 and B2. After this cleaning the system is ready to receive the gas load.

The intermediate storage cylinder should be cooled @77 K by the cryogenic system at the same time that the main storage cylinder is heated @323 K to facilitate the gas transfer from the main cylinder to the intermediate cylinder due to the thermal gradient. When the two temperatures reach their nominal values, the valves that communicate the cylinders are opened and the gas flows naturally from the main cylinder to the intermediate one.

Due to the cryogenic effect, the gas is trapped in the intermediate cylinder and all valves are closed. At this point the heating of the intermediate cylinder is started up to 323 K. The valve communicating this cylinder with the irradiating chamber is opened as is the valve of the target port pressure sensor. The gas is naturally transferred by temperature difference and the pressure is monitored by the supervisory software. The remaining gas in the intermediate cylinder is removed by cryogenic pumping to the main cylinder. At this point the system is ready to initiate irradiation.

3.3. Target Irradiation

During ^{124}Xe irradiation, the temperatures of the water and helium cooling systems should be monitored at all time by the cyclotron operator, therefore in case of sudden changes the irradiation should be stopped immediately. Another parameter that must be constantly monitored is the irradiation chamber internal pressure, which should increase gradually over time with increasing beam current. If a drop in system pressure is observed, the irradiation must be interrupted because a leak must be present.

3.4. End of Bombardment (*EOB*)

At the end of irradiation, wait for 2 minutes until the whole system has returned to room temperature. After this time, all cooling systems can be switched off and the system will be in standby mode for a period of 5 to 6 hours waiting for ^{123}I to form.

3.5. ^{124}Xe Recovery and ^{123}I Withdrawal

After the formation time the ^{123}I remains adhered in powder form to the inner walls of the irradiation chamber, and the ^{124}Xe remaining in the irradiation chamber must be recovered for future irradiation.

This process step is essential for a good outcome of the whole production process. In this step, the vacuum pumps B1 and B2 are turned on again, cleaning the transfer lines through which the ^{124}Xe gas and the water+^{123}I solution circulate.

The first step to be implemented should be the xenon gas recovery for future use as it is very expensive material. To do this, the main storage cylinder must be cooled @ 77 K, then the valves connecting the irradiation chamber to the main cylinder are opened and the gas is naturally transferred due to the thermal gradient between these two points. After the transfer is completed, the valves are closed and the enriched xenon remains trapped inside the main cylinder.

The next step is to extract the ^{123}I that is adhered to the inner walls of the irradiation chamber. For this, the chamber is completely filled with sterile water (approx. 130 ml) and heated @ 343 K and held at this temperature for 20 minutes, this allows that all ^{123}I adhered to the walls to migrate to the water remaining adsorbed thereon. After this time, the cooling water system is turned on to cool the chamber and its contents until the temperature reaches 298 K. At this point, the water+^{123}I solution should be transferred to a hot cell specially prepared for this purpose. The transfer is done by pressurizing the chamber with helium gas. At the end of this process, the step of cleaning and drying can be started.

3.6. Target Port Cleaning and Stand-by Mode

This step requires a long period of time because it is necessary that the moisture left in the system due to the target port washing is removed. The vacuum pump B2 cleans the irradiation chamber and the pipe through which the water+iodine is withdrawn. Likewise, the vacuum pump B1 cleans the Xenon transfer tubing. When the B2 vacuum sensor reaches 5.0×10^{-2} mbar, this part of the system will already be clean. The entire process is complete when the B1 vacuum sensor reaches 1.0×10^{-3} mbar. Since the iodine production is not daily, the system is put into standby mode by filling the irradiation chamber with helium gas up to 2 bar of pressure. This operation has a dual function: to inhibit the entrance of air and humidity in the irradiation chamber and to verify possible leaks in the target port before the next irradiation.

4. EXPERIMENTAL PROCEDURES

The irradiation system tests were done in two stages. In the first one, irradiations were performed using helium as a target element, whose objective is to observe the behavior of the cooling system temperatures (water and helium) and the irradiation chamber pressure as a function of the beam current, because these parameters are decisive for maintaining the integrity of the entire system. Temperatures were measured using K-type thermocouples and the pressure was monitored by means of a strain gage sensor.

In the second stage of the tests, the control and monitoring system was triggered for the ^{124}Xe transfers into the target port irradiation chamber. The pressures used in the tests were 1.1 and 1.5 bar respectively, The results were satisfactory, releasing the system to the next phase, that is, the irradiation.

During irradiation, for a better observation of the interest parameters, the beam current on the target was increased in steps of 5 μA to the maximum value of 70 μA, with an interval of 10 minutes between two steps. The temperatures values of the cooling systems (water and helium) and pressure at the target port were noted and the results are shown in Figures 8 and 9 below.

After the irradiations tests, the system was put into stand-by mode to await the ^{123}I formation. Then, employing again the developed supervisory software, the ^{124}Xe was transferred from the target port to the main storage cylinder. After this step, the supervisory begins the ^{123}I withdrawal procedure and sends it to the hot cell. After this step, the software initiates the cleaning and drying procedures of the target port and the pipes, leaving the system ready for the next irradiation.

5. RESULTS

In the first test of the automated irradiation system, the specific yield (*EOB*) was 5.92 mCi/μAh and in the second one, the specific yield (*EOB*) was 4.63 mCi/μAh. Activity measurements were made employing a CRC-15 Dual PET (Capintec, Inc.)

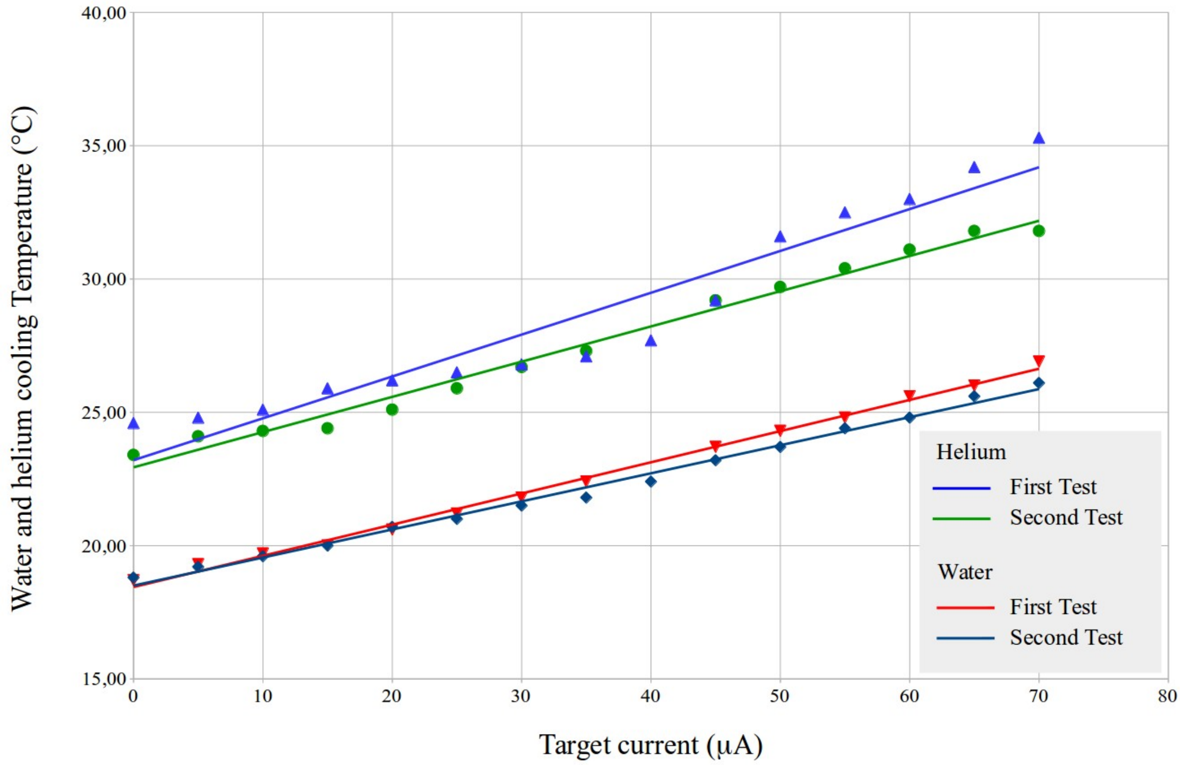

Figure 8: Water and helium cooling temperature as a function of the beam current.

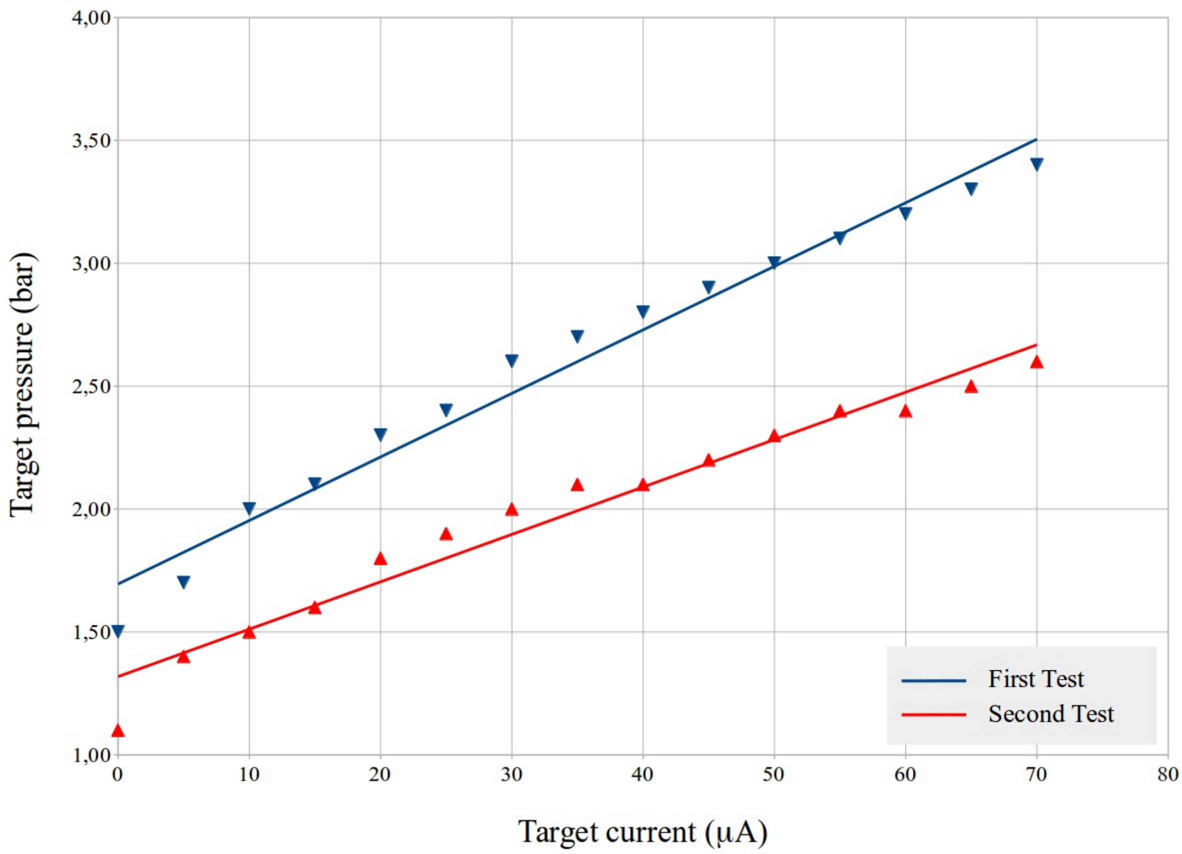

Figure 9: Pressure inside the irradiation chamber as a function of the beam current.

6. CONCLUSIONS

In general, the remotely controlled irradiation system proved to be quite robust and reliable. Adjustments were implemented during testing as needed. Long-term tests should be performed to measure the reliability and stability of the system as a whole.

The results obtained with the tests of performance during the irradiations were satisfactory and within the expected for this type of project. The results showed that the automated system presented in this work is promising so that in a short time it will be used in ^{123}I routine productions by IPEN.

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