EXPERIMENTAL FACILITY FOR NOBLE GAS TRANSPORT IN LIQUID LEAD

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A proposed method for fuel failure detection in lead-cooled fast reactors involves the tagging of the fuel elements with isotopic mixtures of noble gases like xenon and krypton during manufacturing. By inserting inert gases with distinct isotopic composition into fuel pins, operators can trace and identify failed pins through mass spectrometry of the cover gas plenum, enhancing the reactor safety and reliability. However, knowledge gaps remain regarding fission gases transport in lead-cooled fast reactors. To address this, an experimental facility was designed to investigate noble gases migration in molten lead, focusing on the time delay between gas release and its detection in the cover gas.

Keywords: ALFRED, lead-cooled systems, noble gases, tagging gas, reactor safety

1. Introduction

The failure of a fuel element cladding in a nuclear system leads to the release of fission gases into the primary system. Early detection of these volatile elements and precise identification of failed fuel is essential for the safe operation of lead-cooled fast reactors.

In any nuclear system, noble gases such as Kr and Xe are produced through nuclear reactions inside a fuel pin and can escape into the coolant in case of fuel

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cladding defect. Due to their chemical inertness, it is expected that these gases will be promptly and completely released from the molten lead [1].

To enhance defect detection a proposed method involves tagging fuel elements with unique isotopic mixtures of Xe and/or Kr during the manufacturing process [2], [3]. This method involves adding a small amount of inert gas with a unique isotopic composition to all pins of an assembly. During reactor operation, if a pin fails, some of the tagging gas and fission gas escape into the coolant and accumulate in the cover gas plenum. Through the analysis of the cover gas composition using mass spectrometry, operators can precisely identify the isotopic signature of the tagging gas, thereby pinpointing the location of the defective pin. Implementing such a method, operators can mitigate safety risks more effectively, improving overall reactor reliability.

However, there are still gaps in understanding the transport of fission gases in the primary circuit of lead fast reactors. To address these issues, the ongoing ANSELMUS Project in the HORIZON-EURATOM framework [4] aims to investigate the feasibility of detecting failed fuel via cover gas measurements and to assess the impact of noble gas transport on the main circuit of a heavy liquid metals cooled reactor (HLM), including its influence on the time delay between the occurrence of a leak and its detection in the cover gas. RATEN – ICN, actively involved in this project, is focused on studying the migration of noble gases in the cover gas above the liquid lead in a dedicated facility [5].

The Noble Gases Transport in Liquid Lead Experimental Facility (NOGATILL) aims to explore the migration of noble gases (Xe and Kr) in the cover gas above molten lead. Noble gases will be injected into the test vessel and measured online in the cover gas using a Quadrupole Mass Spectrometer (QMS). The duration between the release of a gas bubble and its detection in the cover gas can be examined as a function of gas flow rate.

2. Noble gases transport assessment

Fission gases accumulate in the plenum of the pin-fuel element and they are released into the molten lead only in the event of cladding failure. Early detection and precise identification of failed fuel subassemblies are essential for the safe operation of lead-cooled fast reactors. Accurate identification is decisive as failed fuel assemblies must be promptly removed.

Gas tagging is one of the methods developed for identifying fuel assemblies with minor cladding defects. This technique has been studied extensively for sodium-cooled reactors [6] and has been also investigated for its applicability to the ALFRED reactor [7]. The method involves adding a small amount of inert gas with a unique isotopic composition to all pins of a fuel assembly. During reactor operation, if a pin fails, some of the tagging gas and fission gases escape into the

coolant and accumulate in the cover gas plenum. Subsequently, a mass spectrometric analysis of cover gas samples enables the identification of the failed assembly. This is achieved by measuring the concentration ratios of tagging isotopes and comparing to the initial gas ratios added during fuel pin manufacturing, adjusted for changes due to neutron exposure of the pin [8].

Xenon and krypton are chemically inert with lead and other reactor components making them suitable candidates for tagging. These noble gases are also produced through nuclear reactions within the fuel-pin. Therefore, their effectiveness relies on the equilibrium activity released from the tagging gas exceeding that of the intrinsic fission products produced. The amount of the tagging gas will not exceed 2-3% of the total volume available in the fuel pin leaving enough space for the fission products to build up in the plenum. The maximum volume of tagging gases inserted in the fuel pin gas plenum at standard pressure and temperature (1 bar and 20°C), will range from 2 to 10 Ncm^3 [7]. This results in a tagging gas concentration in the ALFRED cover gas (80 m^3) ranging between 25 and 125 ppb, respectively, assuming the complete release in case of fuel cladding failure.

The inventory of Xe and Kr isotopes produced in a single pin of a fuel assembly (FA) of ALFRED reactor (reference configuration) at the End of Cycle (EOC), was calculated using the FISPACT and MCNPX codes in the framework of the LEADER project [9]. Fig. 1 illustrates the inventory of Kr and Xe produced in a single pin.

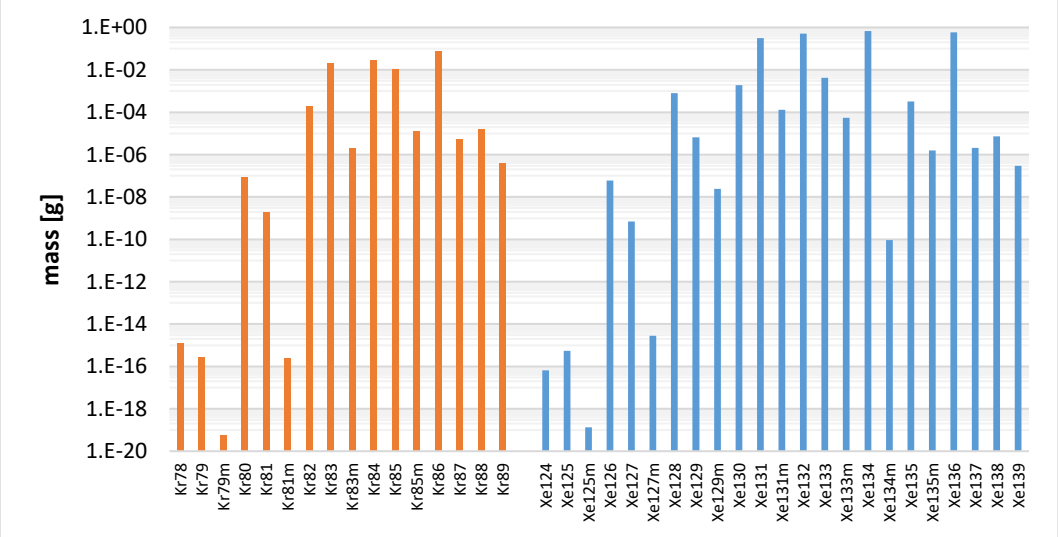
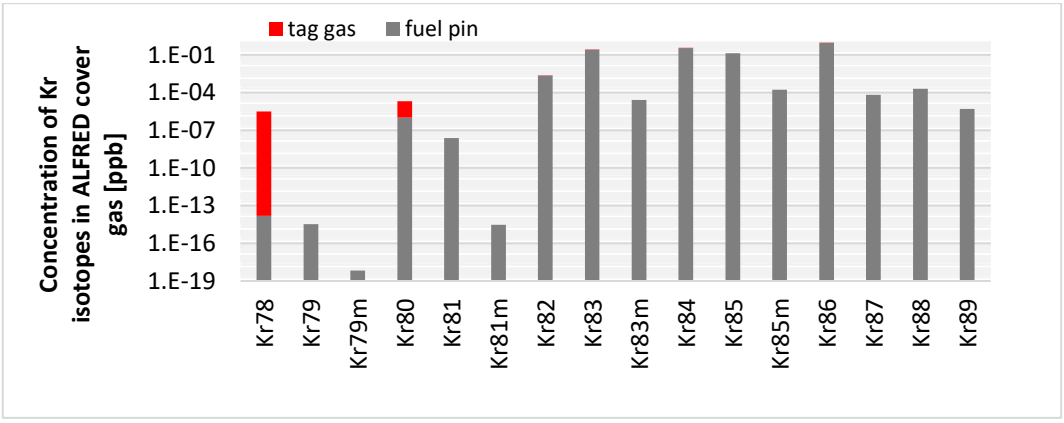


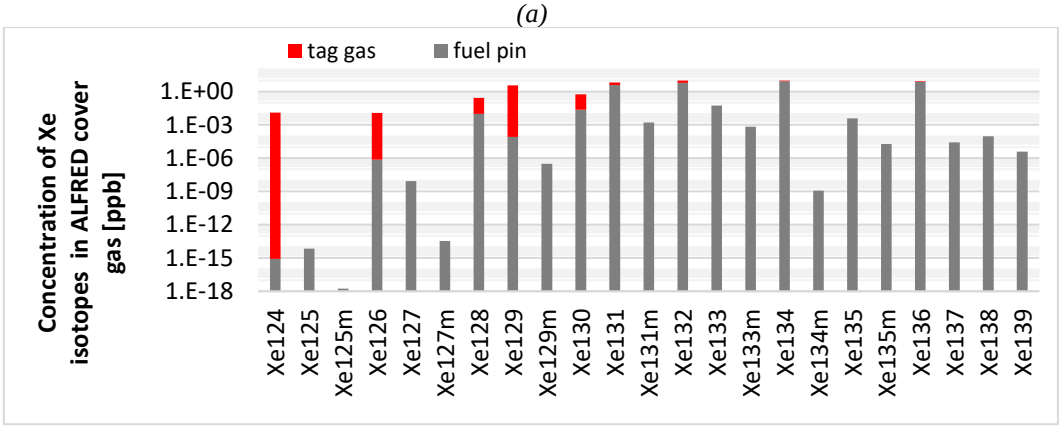
Fig. 1. Inventory of Kr (orange columns) and Xe (blue columns) isotopes formed in a single pin of the FA at EOC

Fig. 2 illustrates the gas concentration in the ALFRED cover gas, assuming complete release of the fission gases produced in a single fuel pin at EOC along with 2 Ncm^3 of tagging gas of natural isotopic composition [10] under 1 bar and 20°C.

Since the concentration of noble gases released into the cover gas is anticipated to be of ppb order or even lower, as depicted in Fig. 2, it is important to select appropriate instrumentation with low and fine detection limit to monitor the noble gas release in the event of fuel cladding failure. One proposed method is to measure the stable isotopes using a quadrupole mass spectrometer or the radioisotopes using gamma-spectrometry as detailed in [7].



(a)



(b)

Fig. 2. The Kr (a) and Xe (b) isotopic concentration in the ALFRED cover gas

Fig. 3 presents the contribution of Xe and Kr radioisotopes to the total activity that could potentially be released in the event of a cladding failure.

The isotopes intended to be used as parent isotopes in the gas tagging process are ^{78}Kr, ^{124}Xe, ^{126}Xe and ^{128}Xe. These isotopes subsequently decay into

^{79}Kr, ^{125}Xe, ^{127}Xe and ^{129m}Xe, respectively. These isotopes are considered suitable options for a fast reactor using MOX fuel [7].

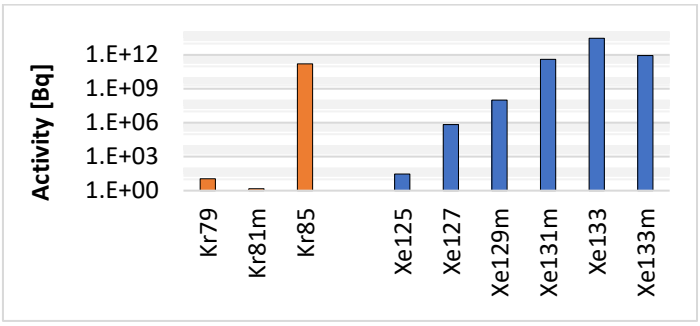
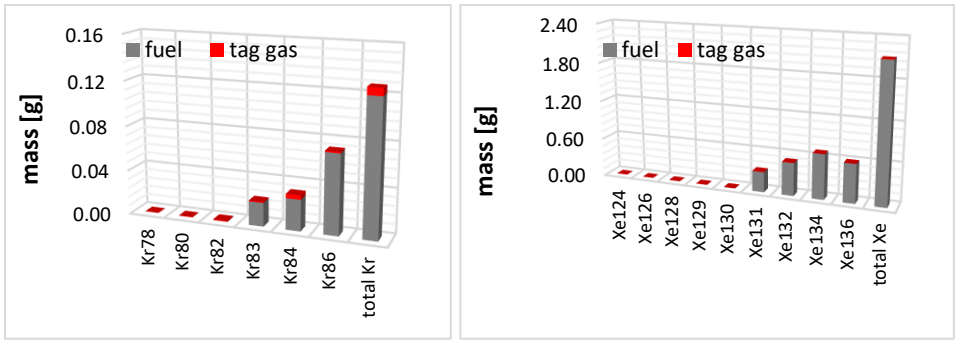
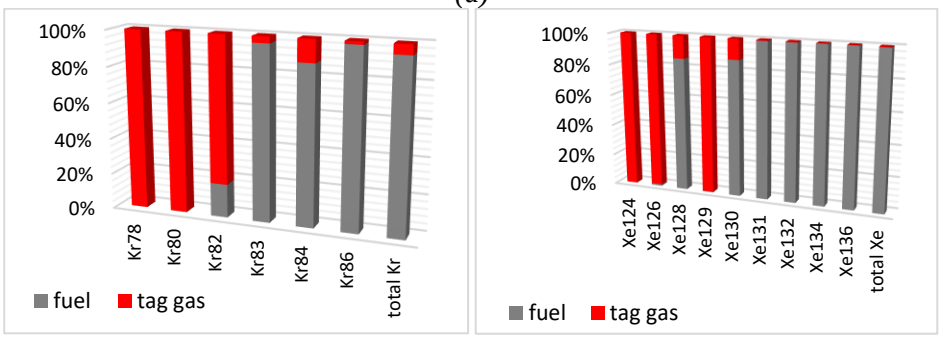


Fig. 3. Activity of Kr (orange columns) and Xe (blue columns) radioisotopes ($T_{1/2} \geq 600$s) accumulated in a single pin of the FA at EOC

In Fig. 4(a) is presented the masses of the Xe and Kr isotopes produced in a single fuel pin along with 2Ncm^3 tagging gas, while Fig. 4(b) shows their relative contribution to the released mass in case of fuel cladding failure.



(a)



(b)

Fig. 4. The masses of and Kr and Xe isotopes produced in a single fuel pin* along with 2Ncm^3 tag gas (a) and each isotope relative contribution (b)

*at EOC and except the radioisotopes

Starting from these assumptions and previous studies, a dedicated experimental facility was designed to investigate the transport of noble gases through a column of molten lead, to establish the capability of mass-spectrometry analysis in determining the duration from the release of a gas bubble to its detection in the cover gas.

3. Experimental facility for noble gases transport in liquid lead

The conceived experimental facility aims to investigate the migration of Xe and Kr into the cover gas above the molten lead. To achieve this objective, noble gases will be injected into the test vessel and monitored in real-time in the cover gas using a quadrupole mass spectrometer. The duration between the release of a gas bubble and its detection in the cover gas can be examined as a function of gas flow rate.

The experimental setup, depicted in Fig. 5, comprises three main components: a melting vessel, a storage vessel, and a test vessel. The vessels are interconnected via a network of pipes and tubes facilitating either the transfer of molten lead between vessels or the circulation of various gases necessary to carry out the experiments. Thus, Argon is used to maintain a gaseous cover above the molten Pb in the vessels, the Ar/H_2 (95%/5%) mixture is needed for O_2 content reduction in lead while the noble gases (Xe and Kr) are to be injected into the test vessel.

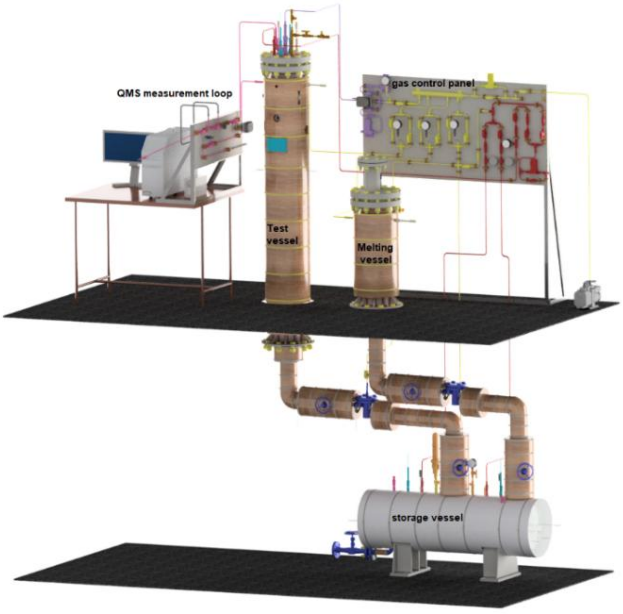


Fig. 5. Noble Gas Transport in Liquid Lead Experimental Facility (NOGATILL)

3.1. Melting vessel

The melting vessel, depicted in Fig. 6, is dedicated to the initial melting of solid lead and transferring the molten lead to the storage vessel. It is a cylindrical, vertically positioned container with a height of 1200 mm and an internal diameter of approximately 200 mm. The solid lead is introduced through the upper flange into a basket that can retain solid impurities contained by lead. The melting vessel can be filled with maximum 3 litres of lead, meaning that the melting and transfer of lead to the storage vessel will be carried out in multiple stages until the final loading of 70 liters of molten lead into the storage vessel is completed.

The melting process is conducted in a controlled argon atmosphere to prevent lead oxidation. The melting temperature is maintained within the range of 350–400°C.

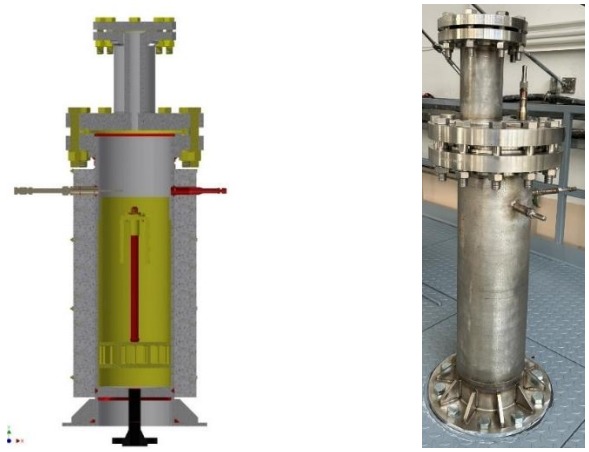


Fig. 6. The melting vessel design (left) and construction (right)

3.2. Storage vessel

The storage vessel has two functional roles: to reduce the concentration of the dissolved oxygen in Pb up to 10^{-6} - 10^{-8} wt% using a 95%Ar/5%H_2 mixture, and to facilitate the transfer of the conditioned lead to the test vessel.

Oxygen is the most critical element in any lead-based system due to its contamination rate and the consequences of solid oxide contamination, which significantly impact the corrosion rate of iron-based structures [11], [12], [13]. Contamination with oxygen occurs during start-up operations, periodic maintenance, and potentially from accidental contaminations, when the atmosphere is nearly saturated with O_2, unlike during normal operation when the oxygen source should be negligible [14], [15].

The optimal range for the concentration of oxygen required for the safe operation of an LFR-type system, ensuring both corrosion protection and non-contamination of the liquid metal, is defined as:

$$C_{O,magnetite} \leq C_O \leq C_{O,sat} \quad (1)$$

where, $C_{O,magnetite}$ is the minimum concentration required for the formation of magnetite (Fe_3O_4) on steel surfaces, C_O represents the concentration of dissolved oxygen in the liquid metal and $C_{O,sat}$ is the maximum limit of O_2 concentration (saturation), [1].

For ALFRED reactor, the strategy regarding coolant chemistry and oxygen control involves maintaining an oxygen concentration significantly lower than saturation [16], as depicted in Fig. 6. Essentially, the oxygen concentration during reactor operation will be kept within the range of 10^{-6} to 10^{-8} wt.% to minimize the possibility of lead oxides formation. The graph illustrates the optimal passivation region for steels within a lead-based system. The same conditions will be fulfilled in the experimental facility presented in this paper.

The oxygen concentration monitoring of the lead within the storage vessel is performed in two points using two potentiometric sensors based on the conductivity of oxygen ions in a solid electrolyte.

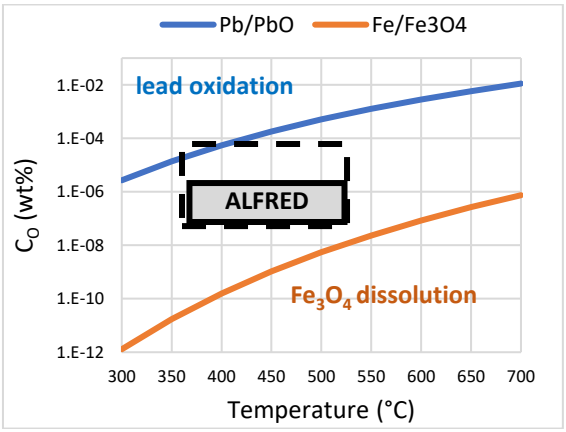


Fig. 6. The control domain of the oxygen concentration in a molten lead nuclear system: the blue line represents the Pb/PbO equilibrium and the saturated oxygen concentration ($C_{O,sat}$) [1]; the orange line corresponds to the Fe/Fe_3O_4 equilibrium and the minimum oxygen concentration ($C_{O,magnetite}$) [16].

The oxygen excess in the lead is removed through a deoxygenation procedure using a gas mixture of Ar and H_2 (Ar 95% + H_2 5%) which is introduced through two penetrations in the storage vessel reaching its base. The Pb oxides react

with H_2 from the gas mixture forming water vapours which reach the cover gas of the storage vessel being further evacuated into the atmosphere.

The storage vessel (Fig. 7) is a cylindrical container having a length of approximately 1300 mm and an inner diameter of about 300 mm, horizontally positioned for operation, the lead volume being of approximately 70L. The operating pressure during lead conditioning will not exceed 1.5 bar, while the maximum pressure value during lead transfer to the test vessel will be of 6 bar. The maximum operating temperature is of 500°C.

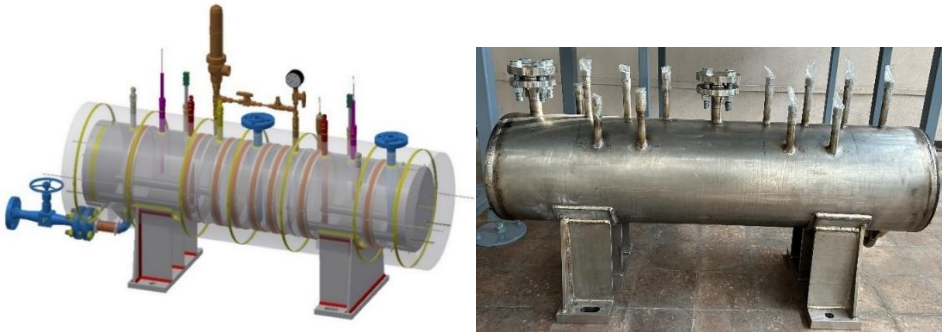


Fig. 7. The storage vessel design (left) and construction (right)

Once the optimal concentration of O_2 is achieved in the molten lead, it will be transferred to the test vessel by pushing it using Ar injected into the storage vessel. Prior to the transfer, the test vessel is preheated and flushed with argon to remove any moisture and prevent the lead from freezing during the transfer process.

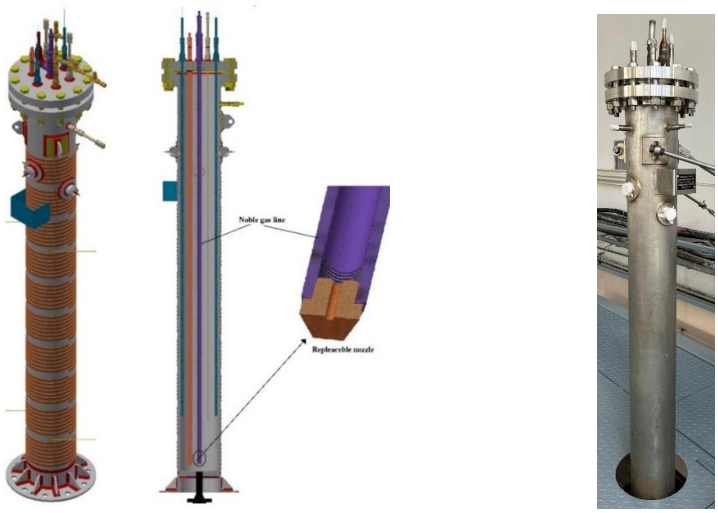
3.2. Test vessel

The gases to be analysed (Xe and Kr) will be introduced through a tube penetrating the vessel lid reaching as close as possible the bottom of the vessel. The tube having a diameter of 0.5 in ends with a replaceable nozzle allowing the adjustment of the orifice size. This setup ensures that the released gases are crossing the molten lead along the whole length of the vessel.

The noble gas injection line is connected to the gas cylinders enabling either the injection of a single gas or a controlled mixture of gases. Moreover, the injection line will be connected to the Ar line providing the necessary pressure to insert the noble gases into the lead. The injection line will be equipped with mass flow controllers allowing the gas/gas mixture injection at flow rates ranging from 0 to 10 ml/min.

The injected noble gases will migrate in the cover gas above the molten metal where they will be measured using a QMS. Once the gas measurements are completed, the noble gases are sent back in the cover gas.

The test vessel (fig. 8) is a cylindrical container with a length of approximately 2000 mm and an inner diameter of about 200 mm, connected through its bottom to the storage vessel allowing the conditioned lead transfer. The free lead level in the test vessel is of 1600 mm – representing 25% of the lead level in ALFRED reactor (calculated from the reactor core to the cover gas). The tests will be performed for temperatures ranging from 400 up to 500°C.

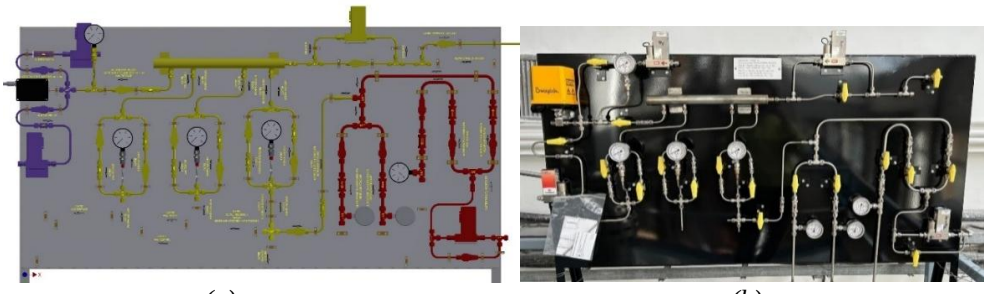


(a) *(b)* *(c)*

Fig. 8. The test vessel: (a) design; (b) cross-section with zoom on the noble gas line injection and nozzle; (c) construction

3.3. The gas system

The gas system encompasses all the lines connecting the vessels ensuring the inert cover gas above the molten lead, the insertion of 95% Ar /5% H_2 mixture into the molten lead, the injection of noble gases into the test vessel as well as the cover gas measurement loop. In Fig. 9 is depicted the gas control panel designed to assure the technical gases required for the operation of the facility. Each system is presented further in detail.



(a) *(b)*

Fig. 9. The gas control panel: (a) designed lines: Ar - yellow; Ar+H_2 mixture - red; Xe and Kr – purple; (b) gas panel construction

3.3.1 Argon supply system

The Argon supply system (yellow lines in fig 9(a)) consists of various components including an Ar gas cylinder, pipelines as well as monitoring and controlling systems.

It allows the formation of the inert cover gas above the molten lead in the vessels and the transfer of the conditioned lead from the storage vessel to the test vessel. It comprises four main lines: cover gas line for the melting vessel, cover gas line for the storage vessel and for pushing the conditioned lead to the test vessel, cover gas line for the test vessel and noble gas transport line to the test vessel.

The Ar pressure is monitored in the cover gas plenum of all three vessels, and the gas flow is controlled using a mass flow controller F201CV from Bronkhorst, calibrated for 1000 ml/min at 10 bar and 20°C [17].

3.3.2. Lead conditioning circuit

The Pb conditioning line (red line in Fig. 9(a)) includes a gas mixture cylinder of 95% Ar /5% H_2, pipelines and monitoring and control systems. It is designed to reduce the O_2 content from lead oxide using H_2 and to produce water vapours that are further removed from the cover gas using the cover gas circulation system.

During lead conditioning the pressure in the cover gas is monitored using the pressure gauges mounted on the gas-mixture lines. The O_2 concentration dissolved in lead is monitored using oxygen-sensors special designed for liquid lead, custom made and provided by Metaprojects Italy [18] and calibrated for high temperature (550°C). The gas mixture flow is controlled using a Bronkhorst F201CV mass flow controller, calibrated for 500 ml/min at 10 bar and 20°C.

This gas mixture circuit consists of two main lines:

- The Pb conditioning line for the storage vessel which splits into two lines penetrating the vessel until close to its bottom.
- The Pb conditioning line for the test vessel to be used in case of accidental contamination with O_2 of the molten lead.

3.3.3. Noble gases injection line

The noble gas injection line (purple line in Fig. 9(a)) allows the insertion of Xe and Kr into the molten lead. It consists off Xe and Kr cylinders, a mixing chamber and mass flow controllers allowing a precise control of the noble gases flow rates. The mass flow controllers F201CV for Kr and Xe are provided by Bronkhorst and both are calibrated for 90ml/min noble gas flow, at 4 bar and 20°C.

The noble gas line injection is equipped with a mixing chamber, a section where the Kr and Xe gases to be injected into the molten Pb are mixed with Ar,

ensuring the necessary pressure to overcome the pressure in the test vessel thus allowing the gas insertion at the bottom of the vessel.

3.4. Cover gas measurement loop

The cover gas measurement loop facilitates real-time monitoring of the chemical composition of the cover gas in the test vessel using a quadrupole mass spectrometer. This loop allows the investigation of the time interval between the release and detection of the gas bubbles as a function of gas flow rate. Additionally, a pump integrated into the loop ensures the required gas flow rate into the spectrometer. After QMS analysis, the gas is returned into the cover gas. Continuous introduction of the gas into the mass spectrometer at low flow rates enables the detection of any change of its chemical composition in less than 0.5 seconds.

4. Method of analysis and experimental campaign

The measurement of the isotopic composition of the cover gas using a quadrupole mass spectrometer for residual gas analysis, type SRS QMS 200, enables the identification of the evaporated isotopes. Operating under vacuum conditions, the spectrometer absorbs the gas for analysis and maintains it under vacuum. The gas is continuously introduced into the analyser at low flow rates (a few millilitres per minute), making it ideal for real-time measurements. The system allows for rapid data collection providing the acquisition of a complete spectrum in less than one minute [19]. This method has been successfully used in monitoring the composition of fission gases released from CANDU-type fuel and in obtaining the Xe/Kr isotopic ratios [20].

The experimental campaign foreseen in NOGATIL facility can begin once the oxygen concentration in the molten lead is within the established range, and the temperature of the liquid metal is raised to 480-500°C, according to the normal operating conditions of the ALFRED reactor. The chemical composition of the cover gas in the test vessel is measured by recirculating the gas through the QMS measuring loop. Initial monitoring determines the cover gas pressure and composition prior to noble gas injection.

The experimental tests are designed to simulate various scenarios of fission gas release in the event of a failure in the fuel cladding, and include two types of injections:

- ***Single injection***, performed by introducing a known gas volume, between 1 and 10 ml, in a single step. This type of test mimics the conditions of a total and immediate release of fission gases accumulated in the gaseous plenum of the pin, as would occur in the case of a major cladding failure;

- ***Continuous gas flow***, at controlled rates between 1 and 10 ml/min. This scenario simulates the gradual release of fission gases, as would occur in the case of a fissure- or pore-type failure in the fuel cladding.

The noble gas injection can be performed using a single element, such as Kr or Xe, or mixtures of these gases, in different ratios.

After gas injection, the sample release time and the moment when changes in the gas's chemical composition are detected are recorded and monitored in real time. The cover gas in the test vessel is continuously analyzed for at least 20 minutes to ensure the system reaches equilibrium.

The experimental data obtained will significantly contribute to the validation of safety systems for liquid heavy metal-cooled reactors, improving the early detection capability of failures occurring in the fuel cladding.

5. Conclusions

Detecting fuel cladding failure is essential in ensuring the reactor safety. Noble gases produced as by-products of nuclear reactions and accumulated in the fuel pin can serve as important indicators of such defects. Techniques like gas tagging which consists in the insertion of inert gases (e.g., Xe/Kr) with distinct isotopic compositions into the fuel pins can help to pinpoint the failed assemblies.

In this aim, a dedicated experimental facility was designed to investigate the transport of tagging gases through a column of molten lead, aiming to determine the duration from the release of a bubble gas to its detection in the cover gas. Real-time gas analysis using quadrupole mass spectrometry allows for rapid and accurate detection of the tagging gases.

The experimental facility detailed in this paper is currently under construction and is projected to be completed by April 2025. Functional and tightness tests will follow over the subsequent four months to ensure full operational readiness. Noble gas detection experiments within the test vessel are scheduled to commence in the third quarter of 2025.

Acknowledgements

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