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ScienceDirect
Journal of Nuclear Materials 356 (2006) 264–272
journal of nuclear materials
www.elsevier.com/locate/jnucmat

ENEA experience in LBE technology

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Abstract

Since 1999, several facilities have been operated with flowing LBE in the frame of ADS R&D activities at CR ENEA Brasimone. The experimental activities have gone through thermal-hydraulics, heat exchange, science of materials, qualification of components and operational procedures. Thanks to the performed work, a large amount of observations were made, and experience was gained. This paper is focused on the survey of these experiences gained, in order to point out advantages and disadvantages of the LBE technologies. In particular, experience on plant components, measurement instrumentations and operational procedures will be pointed out.

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1. Introduction

Presently proposed accelerator-driven systems (ADS) are subcritical nuclear reactors in which the fission reaction chain is maintained in steady conditions by the injection of neutrons generated in a target material through the spallation reaction by a high energy proton beam produced in an external accelerator. The neutronic characteristics of the reactor are conceived as to efficiently incinerate long-lived fission products by transmutation, in order to reduce the need for very long term geological disposal of nuclear wastes produced in the nuclear reactor fuel cycle [1–3].

From the beginning of the most important research programs on these systems, the lead–bismuth eutectic (LBE) alloy was proposed as one of the most promising target materials as well as coolant for the reactor. Then, since 1999, several small devices as well as large loops and facilities have been operated at the CR ENEA Brasimone, to perform experimental activities aimed to investigate thermal-hydraulics, heat exchange, science of materials, qualification of components and operational procedures topics related to the use of LBE.

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doi:10.1016/j.jnucmat.2006.05.020

2. Experimental facilities

2.1. The LECOR loop

The LECOR loop (lead corrosion) is a non-isothermal, eight shaped forced lead–bismuth loop, which consists of a surge tank, a mechanical pump, a heat exchanger, a heater, three test sections, an air cooler and a storage tank with 600 l liquid lead–bismuth eutectic (LBE). An oxygen sensor was installed close to the test sections to monitor the oxygen activity in LBE during the operation of the loop. The detail description of the LECOR loop is given in Ref. [12].

2.2. The CHEOPE III loop

The CHEOPE loop (chemistry and operations) is installed in the ENEA Brasimone Centre to evaluate the operation conditions for liquid metal. The CHEOPE III is one part of a multipurpose facility, which was used to perform the corrosion test in a controlled oxygen environment (with high oxygen activity comparing to that in the LECOR loop). The structure of this loop is similar to the LECOR loop. It consists of a storage tank filled with 400 l of LBE, a test section and an oxygen sensor.

In Table 1 the technical parameters of both loops are summarized.

2.3. The CIRCE facility

CIRCE (circulation experiment) [4–6] is a large-scale test facility designed for studying key operating principles of the 80 MW experimental accelerator-driven system (XADS). It basically consists of a reduced diameter (1:5 the XADS vessel diameter), full-height, cylindrical vessel (Main Vessel S100) filled with about 70 of tons molten LBE with argon cover gas and recirculation system, LBE heating and cooling systems, and auxiliary equipment for eutectic circulation [4,5]. Dedicated test section can be housed in the main vessel. The facility is completed by a LBE storage tank, by a small LBE transfer tank and by a data acquisition system. The facility is designed to perform large-scale experiments to investigate the thermal-hydraulic, chemical and mechanical issues related to the development of the LBE-cooled XADS in a pool configuration. The main parameters of CIRCE are given in Table 2.

Table 1
LECOR and CHEOPE main working parameters

Parameters	LECOR loop	CHEOPE III loop
Temperature in the test section (°C)	400	400
Temperature in the cold part (°C)	300	300
Test time (h)	1500, 4500	1500, 3000, 4500
Velocity in the test section (m/s)	1	1
Oxygen concentration (wt%)	10^{-10}–10^{-8}	10^{-6}–10^{-5}

Table 2
CIRCE main vessel parameters

Outside diameter	1200 mm
Wall thickness	15 mm
Height	8500 mm
LBE inventory	70 tons
Operating temperature range	200–550 °C

3. Plant components

3.1. Standard component available on market

The experimental facilities have been built mounting the standard components available in the specialized scientific and industrial markets in order to reduce the costs of the research activities and the time for the implementation of the facilities; in this way was possible to improve the reliability and the repeatability of the tests.

For example, the measuring instrumentation includes calibrated K-type thermocouples, electromagnetic and vortex flow meters and differential pressure transducers, which are commonly utilized in standard industrial applications; the data acquisition system that consists of a personal computer, a data acquisition board and several conditioning modules, is the same kind of the other ones installed in several facilities.

3.2. Mechanical pumps

The pumping system, at the first moment, had some problems related to the pump chosen. It was a Novatome (Fr) electro magnetic pump. The deposition of LBE oxides over the internal surface of the pipe compromised the functioning of the pump, downgrading its already low efficiency (~5%).

In order to solve this problem, two mechanical pumps (see Table 3) were installed on the LECOR

Table 3
Pumps working conditions main parameters

Loop	Flow (m^3/h)	Load/lift (bar)	Working temperature (°C)
LECOR	4	5	300
CHEOPE	1	3	300

and CHEOPE loops. These pumps are of submerged vertical axis kind. Their electrical motor should be segregated away from the LBE.

In the case of the LECOR pump, the perfect insulation between the impeller vessel and the electric motor is guaranteed by a rotating seal (Chesterton) fitted on the pump tree. Even if the Chesterton seal is expensive and needs to be cooled (working temperature <200 °C) with a dedicated oil circuit, this is a very reliable solution that makes the service of the electrical components of the motor very comfortable.

In order to avoid the contamination of LBE due to oil release, it is very important to check the rotating seal periodically.

The solution adopted in the CHEOPE loop is different. The whole motor was sealed to the flange of the pump vessel. In this case a cooling system is needed for the electric parts and the service is uneasier than in the previous case.

3.3. Flow meters

Two different kind of flow meters were mounted on the facilities: some Novatome electromagnetic induction flow meters (EIFM) and some Foxboro vortex flow meters. Table 4 summarises the operational conditions of the instrumentation.

The EIFMs were calibrated, on theoretical calculations, before fitting on to the facilities. During the experimental campaigns the EIFMs gave an incoherent output signal, that tent to zero with time. Only the ∅ 1″ put on the cold leg of the LECOR loop still works correctly. The benchmark of the output signal is made through a thermal balance.

After the conclusion of the TECLA project all FMs were dismounted in order to service them. The measurement behaviour of the EIFMs was caused by the growth of an oxidized layer on the internal surface of the piping. This plugging layer worked as an insulator, lowering the measure capability of the instrumentation, until the complete annihilation. Only one EIFM, (∅ 1″), is still working. This depends on two different causes; first of all its diameter is wider than the other diameters, so the plugging layer does not cover the whole surface, secondly it is working in a reducing environment which slows the growth of the oxide layer.

3.4. Pressure transducer

In order to measure the pressure variations, three different transducers were mounted on the loops CHEOPE and LECOR. The functional principle of these three instruments is quite the same: a diaphragm faces directly the LBE. On the other hand it is also in contact with a filling fluid by which the variations of pressure are transmitted to the transducer. The difference among them is given by the membrane dimensions and their surface shape (polish vs. corrugated). Table 5 summarises the operational conditions of the instrumentation.

Both Kulite and Gefran pressure transducers went out of work after few hours. This was caused by oxides plugs which covered entirely their polish diaphragm (∅ 5 mm).

In contrast to this the Rosemount one is still working thanks to its wider membrane (∅ 95 mm). Moreover, the corrugated shape minimizes the blocking effect of the oxides.

3.5. Prototypical components

In order to realize and to operate the facilities it was necessary to project, construct and calibrate a new kind of measuring instruments which had been never used in standard applications. These proto-

Table 4
Flow meters working conditions main parameters

	Technical data	Measure range	Facility	Working temperature (°C)	Time (h)
EIFM	∅ 1/2″	0–4.5 m^3/h	CHEOPE	300	1000
	∅ 1/2″	0–4.5 m^3/h	LECOR	400	5000
	∅ 1″	0–11.25 m^3/h	LECOR	300	5000
Vortex	∅ 1″	0.10–3.6 l/s	CHEOPE	300	4000

Table 5
Pressure transducers working conditions main parameters

	Technical data (∅ membrane) (mm)	Measure range	Facility	Working temperature (°C)	Time of work (h)
Kulite (UK)	5	0–7 bar	CHEOPE	300	~1000
Gefran (I)	5	0–0.35 bar	LECOR	300	~ 1000
Rosemount (USA)	95	0–276 kPa 0–240 kPa 0–2070 kPa	CHEOPE	300	~5000

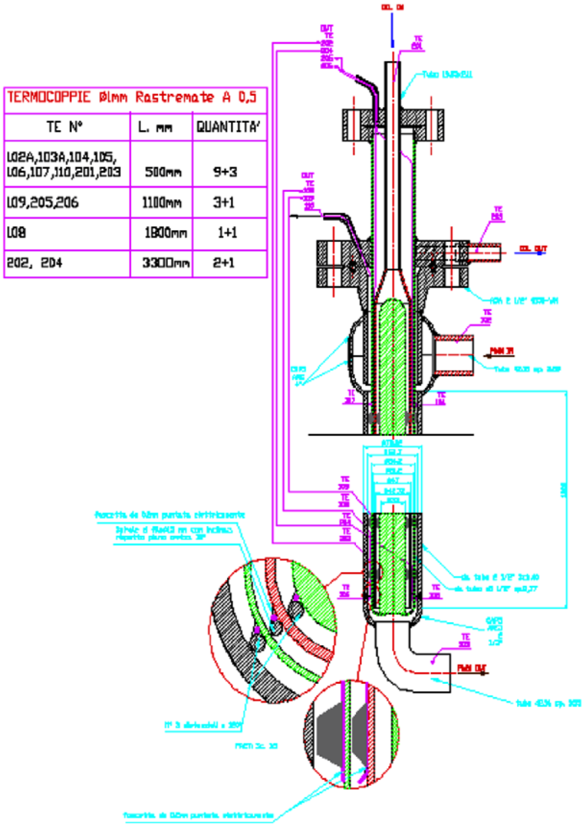
typical components, described in the next paragraph, are inspired to standard components, but are modified to improve their measure magnitude, reliability and sensitivity in applications involving LBE.

3.6. Pin cooler

In the frame of the Megapie Project, a prototypical configuration for intermediate heat exchanger between LBE and organic oil Bayer Diphyl THT was studied in ENEA. The configuration proposed by PSI was a bayonet heat exchanger named cooling pin. Its scheme is shown in Fig. 1. The design contains some important features:

1. The bayonet pattern allows the differential expansion of steel parts without generating sensible thermal stresses.
2. The mechanical mount allows an easy intervention for inspections and repairs, being based only on flanges connection/disconnection.
3. The thermocouple mounting allows a redundant monitoring of the thermal behaviour.
4. Helix coiled wires enhance the thermal exchange per unit length of the component at price of a larger pressure drop.
5. A special LBE intake region with calm-down chamber and thermal shield was designed.
6. Thin annular channels were obtained to enhance thermal exchange by raising the fluid velocities.

The experimental positioning of the thermocouples allows the calculation of the mean logarithmic temperature difference along the whole pin. This was done taking into account the LBE (inlet and outlet) bulk temperatures and the corresponding wall temperatures taken on the LBE side. The thermocouples ($D = 0.5$ mm) were fixed to the wall and covered by a metal sheet (0.1 mm thickness) confining it on the wall side of the boundary layer. The exchanged power value has been obtained by a thermal balance between inlet and outlet enthalpy of oil through the pin. It was calculated measuring the



TERMOCOPPIE Ø1mm Rastremate A 0,5		
TE N°	L. mm	QUANTITA'
102A,103A,104,105, 106,107,110,201,203	500mm	9+3
109,205,206	1100mm	3+1
108	1800mm	1+1
202, 204	3300mm	2+1

Fig. 1. Scheme of the cooling pin system.

following data on the oil side: mass flow-rate, inlet–outlet differential temperature and specific heat.

The specific heats, as well as the other physical quantities, were computed as a function of the mean temperature for both fluids. For each test we calculated the convection coefficient of LBE applying the formula of mean logarithmic temperature difference between LBE temperature and wall temperature at the ends of the instrumented length. The final results were averaged for the whole duration of each single test. Each test can be considered stationary in the sense that the local temperatures did not vary considerably. The experimental results ranged between 10000 and 11000 W/m^2 °C for LBE Reynolds numbers in between 15000 and 18000. The results of the global heat exchange coefficient show that it is mainly influenced by the oil coefficient which is the lowest one among the three contributions (oil convection, steel conduction and LBE convection). For this reason, the global heat exchange coefficient, which ranges between 1100 and 1700 W/m^2 °C, can be linearly related to the oil-side number of Reynolds power 0.8.

The oil-side spiral diverts of about 30° of the direction of the oil flow. This device enhances the thermal exchanges at the oil side, but introduces two additional reasons for pressure drops: increase of the local velocity modulus and continuous change of direction of the velocity vector. According to the experiments, a total pressure drop of 1926 mbar has been found, oil side, at 0.17 m/s axial speed through an annular channel of 51.2 mm external diameter and 47 mm internal diameter.

3.7. Oxygen sensors

The monitoring of dissolved oxygen is also a fundamental tool in the field of metal structures protection. Keeping oxygen in a controlled range of concentration, i.e. in this specific case $C_{O(MgO)} < C_O < C_{O(PbO)}$, induces the formation of oxides layers over the surfaces of the containing steels, protecting them against corrosion phenomena. Due to this twofold aspect of dissolved oxygen, its concentration monitoring becomes of primary importance.

The active oxygen control system process is performed by separate addition of H_2 and O_2 and the use of a getter (such as Mg), while the monitoring of the oxygen concentration is performed by Russians sensors, produced at IPPE, Obnisk. The only effective dissolved oxygen monitoring systems for liquid metal environments as LBE are online

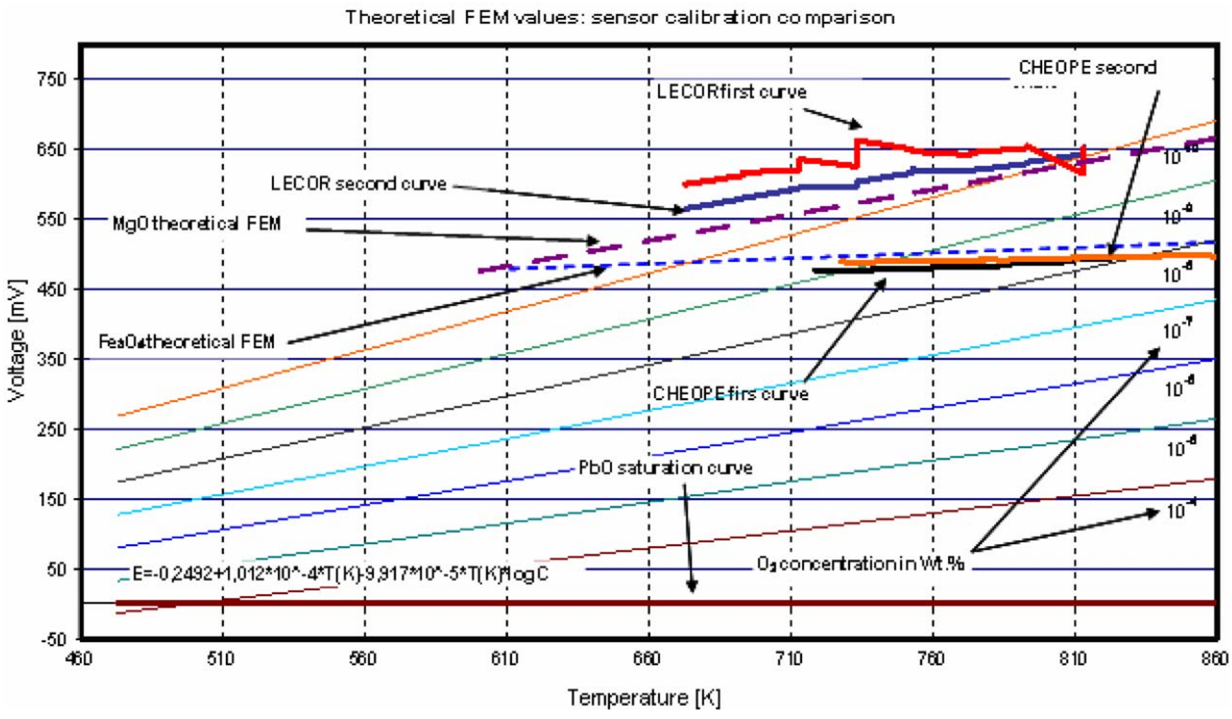


Fig. 2. Sensor calibration curves: voltge [mV] vs. temperature [K].

electrochemical oxygen probes, conceived as galvanic cells in which an yttria–zirconia doped ceramic elements acts as a porous sect. In this simplified system, the liquid eutectic represents the anode, the reference systems the cathode. A ceramic electrolyte, as said consisting of yttria stabilized zirconia, can be permeated by the oxygen ions, establishing a ionic current that is proportional to the oxygen partial pressure, and so to its activity in the liquid eutectic.

EMF is measured by means of a high impedance voltmeter; the working electrode and the electric wires are usually made of molybdenum or stainless steel.

According to the NERNST relations the EMF measured is a direct, online indicator of the dissolved oxygen concentration. A calibration system of these kind of sensors has been developed too, by means of a stagnant vessel with controlled LBE conditions (Fig. 2).

3.8. Venturi-Nozzle flow meter for large-scale experiments

Since the thermal-hydraulic large-scale experiments planned at the ENEA Brasimone Centre were designed to be carried out in a wide range of flow rates, a dedicated Venturi-Nozzle flow meter was manufactured. Of course, the Venturi-Nozzle flow meters are well known devices, but none were used before to perform measurement in LBE and, furthermore, in a range of flow rates up to 350 kg/s.

Such flow meter has been supplied by the Euromisure s.r.l. (I), together with the calculated theoretical characteristic equation. So, before its use in the circulation experiments, a dedicated experimental campaign was carried out for its qualification. The results obtained by the tests performed, allowed to verify the theoretical curve supplied by the manufacturer, that presents a very good agreement with the experimental data. In fact, since the flow-rate range is restricted to 100–350 kg/s, the uncertainty showed is lower than ±3%. Moreover, in order to extend the range of flow rates at lower values with a similar order of accuracy, an experimental equation obtained by best fitting the experimental data has been produced too. Using this best-fit equation, it is possible to perform flow-rate measurement with an uncertainty of ±3% in the range of 40–350 kg/s. Of course, the measurement is not reliable at flow rates lower than 40 kg/s, due to the intrinsic loss of sensibility characteristic of this class of instruments [10,11].

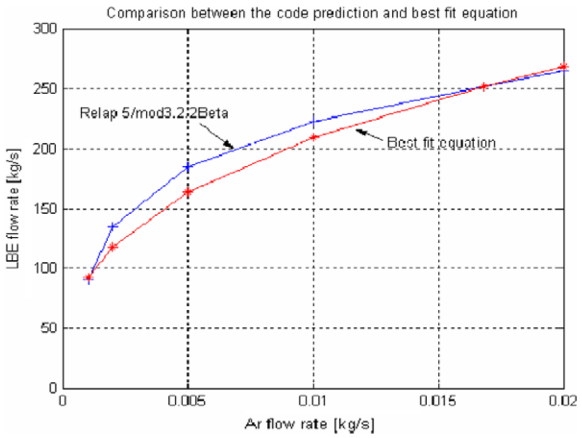


Fig. 3. Comparison between the correlation obtained the best fit of the experimental data and the pre-test calculations.

The Venturi-Nozzle flow meter has been already used during the experimental campaign aimed to investigate the possibility to enhance the LBE circulation in a pool by gas injection, showing good performances. Then, it will be employed during the next integral experiment planned on CIRCE in the frame of the IP EUROTRANS.

In Fig. 3 the comparison between the correlation obtained by best fitting of the experimental data and the pre-test calculations is shown.

4. Plant procedures and special techniques

4.1. Oxygen control and LBE purification procedures

Together with the above described oxygen concentration monitoring, several LBE dissolved non-metallic elements control methods have been implemented. Those methods are resumed in the following list:

1. *LBE solid ingots pre-treatment:* A simple mechanical sanding of the ingots has been found extremely effective in eliminating natural oxides.
2. *Mechanical filtering:* The use of Poral filters during the filling procedure was effective in blocking macroscopic slags. The mechanical filter put in the LECOR filling system did not plug in 6000 h of loop exercise.
3. *Adsorption filtering:* A hot trap with a fiberglass filter has been implemented in the CHEOPE III facility, revealing effectiveness in trapping small slags.

(4) *LBE chemical conditioning:* The concentration of oxygen in the LECOR loop was lowered down to 10^{-8} wt%, by means of Ar/H_2 gas mixture bubbling and the addiction of a stoichiometric amount of solid Mg. Due to its thermodynamic properties, Mg acted effectively as Oxygen getter; gas bubbling demonstrated its effectiveness as a reducing medium. The formed vapour was monitored by an external hygrometer.

4.2. LBE freezing procedure

One of the main risk about the freezing of LBE inside the liquid metal containers is the rupture of the experimental vessels when the frozen LBE starts to expand by recrystallization. A study of the parameters affecting the stress level in the vessel produced by expansion of the solidified LBE was performed in [12,13,14]. The eutectic mixture recrystallization is the key process that must be understood in all its aspects in order to avoid harmful and dangerous events in LBE vessels. A phenomenological interpretation has been suggested as a result of laboratory activities. Recrystallization is a phenomenon that takes place at the level of crystal grain atoms, passing from a state of non-equilibrium towards a state of equilibrium. In adjacent grains different phases are segregated; these form crystal cells with different inter-atomic distances. When temperature varies with time, the atoms migrate through the grain boundaries, passing from one crystal form to another. Due to the atoms migration, during freezing, the solid alloy gets richer in γ phase and, with time, at a generic temperature T lower than T_f (the melting/freezing temperature), the excess γ phase precipitates. The γ phase is richer in Bi than the β phase, and bismuth (unlike lead) expands on solidification. So the γ phase precipitation and the atoms' migration from grain to grain (i.e. recrystallization) tends to generate a volume increase. The stress analysis shows that the field of stresses in a LBE containing vessel is mainly affected by the height of the solid LBE level, the degree of expansion of the contained alloy, the yield stress of the solid LBE the strain rate and the cooling rate.

The height of the solid LBE in the container is a significant parameter which needs to be accurately chosen to avoid over-stressing of the vessel. For this reason the preferred storage tanks have horizontal axis and are filled at less than half of their capacity. For vertical tanks it is advisable to design the LBE height in such a way to respect a minimum ratio of roughly 1/2 between diameter and LBE height. In case of higher tank height the freezing action can be performed in batches. This procedure consists of: (1) start freezing from the lowest layer of appropriate length, while keeping the rest of the LBE liquid; (2) maintain such conditions for a prescribed time (roughly one week) to allow enough expansion; and (3) proceed with the higher layers, so that the liquid volume resting above the level of the solid substrate is free to expand towards the top of the vessel without any constraint.

The LBE yield strength is also a very important parameter. It was shown that increasing the solid LBE temperature, a significant reduction of LBE yield stress takes place, thus reducing the associated wall stress. The current interpretation is that a higher yield stress means that the internal forces needed to extrude axially the material in a plastic flow are higher and the load on the containment structures must be consequently raised. At 110 °C temperature and 5E−06 s^{-1} strain rate, LBE shows a yield stress as low as 3.0 MPa instead of 25 MPa observed at 20 °C [14]. According to experimental results in [15], the LBE yield stress turned out to be a function of temperature, age and strain rate. The LBE yield stress could therefore provide a means of reducing the stress level in the walls by controlling temperature and time of the solidified LBE.

4.3. Level, pressure and differential pressure measurement technique based on bubble tubes

As previously discussed, the measurement of pressure and levels in loops and small vessels is performed using standard instrumentation. However, in the case of large-scale facility, where pool type system is simulated, the use of standard instrumentation can be difficult, mainly due to the thermal and chemical interactions between the LBE and the measurement devices. For this reason, a simple measurement technique based on 'bubble tubes' has been tested in a dedicated device and finally applied to the CIRCE facility. In fact, bubble tubes injecting argon below the molten metal level, have been installed to transfer pressure signals from the LBE alloy to differential pressure cells operating with gas at room temperature. In such a way, level, pressure and differential pressure measurement are performed in the CIRCE pool. This measuring system has various advantages: (1) it includes only a few components, which are all of standard design; (2) since there is no material included to separate the gas region from the melt alloy, there is no additional problem of compatibility with the coolant; and (3) the pressure transducers are located sufficiently apart from the molten coolant, as to be preserved from any possible thermal or chemical interaction with it.

Indeed, the possible drawbacks of this technique, that may come from its intrusive character (mainly consequent to the injection of argon in the coolant which might disturb the flow), and from possible delays in the transmission of the pressure signals along the injection gas lines, have been investigated. The results obtained during the assessment of this technique indicate a good performance under steady conditions, with a reasonable accuracy, showing an error in the order of 2 mbar.

Concerning the availability of the technique, an initial waiting time before data acquisition is recommended to allow the measurement system to settle to asymptotic behaviour. Of course, under transient conditions, in particular positive pressure transient, the loss of a reliable measurement signal can occur, since the gas injected is a compressible medium. However, the problem can be solved using a theoretical model based on standard balance equations, in order to compensate and to correct the output signal [7,8].

In Fig. 4 the assembly drawing of the apparatus is shown. It was used for the qualification of the measurement technique based on bubble tubes.

4.4. Pumping technique based on gas injection: gas lifting

The possibility to enhance the circulation of LBE by gas injection is considered of primary importance for the assessment of the feasibility and operability of an ADS prototype. In fact, this technique will allow the primary coolant circulation with a plant simplification in respect to the use of mechanical pumps. Because of the reliability of this technique was never been proved before on significant scale, a dedicated experimental campaign has been carried out on the CIRCE facility.

The tests have been performed under isothermal condition at 200 °C [9]. In fact, this temperature is the cold shutdown temperature in the Ansaldo XADS concept, and was considered as reference temperature during the pre-test analysis performed by Ansaldo using the RELAP5/mod3.2.2 Beta code (Fig. 3).

The results obtained showed that for Ar flow rates injected higher than 1 N l/s is possible to rise a steady flow of LBE trough the adopted test section, that was able to reproduce the velocity fields expected for the reference ADS plant, in the range from 100 to 250 kg/s. A relation between the

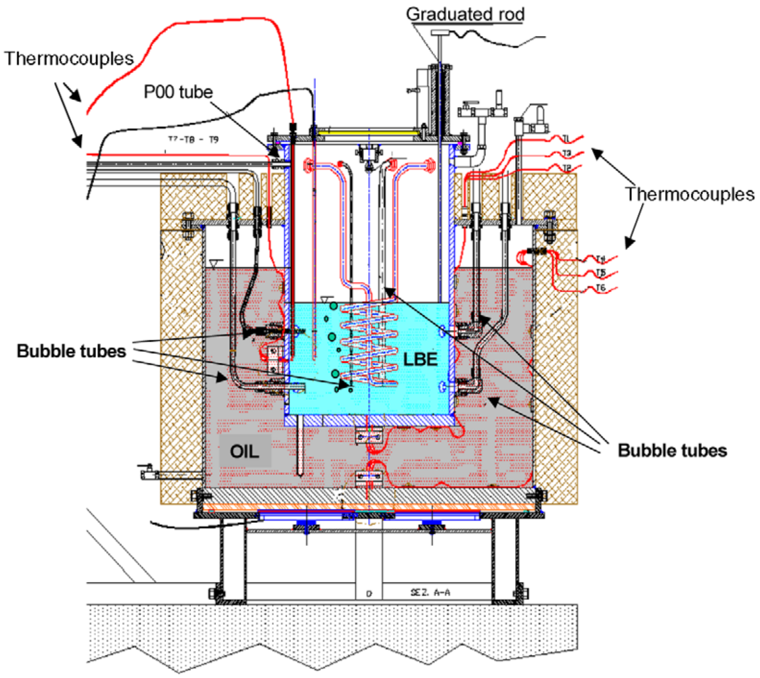


Fig. 4. Assembly drawing of the apparatus used for the qualification of the measurement technique based on bubble tubes.

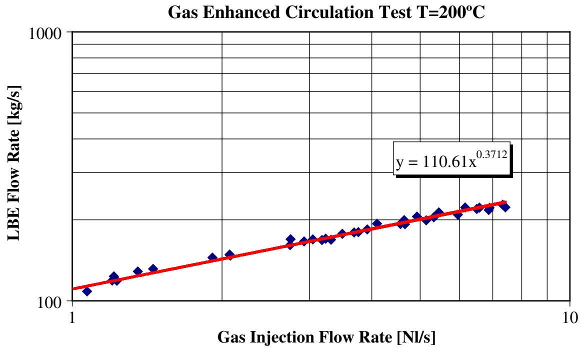


Fig. 5. LBE entrained flow rate vs. Ar injected flow rate, as resulted from the performed tests at 200 °C.

pumped liquid metal flow rate and the injected gas flow has been carried out by best fit of experimental data. This relation follows a power law, and a theoretical justification of the result has been carried out too. Moreover, The Relap5/mod3.2.2 Beta code in the version developed by Ansaldo seems to be in a satisfactory agreement with the experimental data.

Tests to complete the characterization of this technique are now ongoing on the CIRCE facility at different temperatures (up to 320 °C). The results will be used to point out the relation between the void fraction and the pumped flow rate, and to qualify this technique in terms of pumping efficiency.

In Fig. 5 LBE entrained flow rate vs. Ar injected flow rate is presented, as resulted from the performed tests at 200 °C.

5. Future applications

The CR ENEA Brasimone will take part at the Integrated Project Eurotrans [16] with the objects to reproduce, in the CIRCE large-scale facility, the natural and forced circulation of a sector of the primary LBE coolant in the reactor pool with a 750 kW heat source, a cold sink and a secondary coolant. When these tests will be running, it will be possible to obtain information about the operating and incidental transients, to check the mass transfer effect and validate the nodalization parameters for thermal-hydraulics system computer code for steady state and transient analyses of the experimental data.

Acknowledgements

The authors want to thank all the technical personnel of the CR Brasimone – ENEA, for their precious work and help.

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