

The Jules Horowitz Reactor MOLY system. Towards a concept proposal according a large Molybdenum production capabilities.

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Abstract

To ensure the radioisotopes production for the medical applications constitutes a strong stake for our present societies. To guarantee the adapted implementation and the operability of facilities allowing producing continuous and sufficient quantities at the international level require the anticipated and precise definition of experimental irradiations systems regarding to the medical needs.

In this context, the Jules Horowitz Reactor (JHR), which is in the construction phase at the CEA Cadarache (South-east of France) aims at offering an important capacity of radioisotopes production for the medical issues (25 up to 50% of the European needs).

This facility, which is a 100MWth MTR type (Material Testing Reactor) is devoted to carry out fuels and materials irradiations in order to support the actual and future Nuclear power plant programs. It has also been designed to produce MOLY (Mo-99) type and other radioisotopes for the medical sector.

The studies of JHR MOLY devices are performed through collaboration between the CEA and NRG institutes. A preliminary study of the JHR MOLY circuit has been performed using the feedback from the OSIRIS reactor located in Saclay in France and from the HFR reactor located in Petten in the Netherlands. These two reactors assure a significant part of the MOLY production in Europe.

NRG, which operates the HFR reactor has a significant experience in the field of technology interesting and applicable for JHR issue (as cooling systems, irradiation facilities,...).

After a short description of the characteristics of the Jules Horowitz reactor, this paper describes the experience available on the design of irradiation circuits of MOLY type.

In the studies, different concepts of circuits are analyzed taking into account various criteria (operation, availability, flexibility, safety,...). These analyses conclude to a reference architecture proposal of MOLY production systems in the JHR facility environment. This architecture will be presented.

The following stage of these studies will correspond in 2010 to the conceptual design studies of the MOLY circuit and devices for the JHR facility.

Keywords:

Jules Horowitz Reactor, radioisotope, MOLY devices, Molybdenum production

1. Introduction:

The production of Molybdenum 99 (Mo-99) radioisotopes is important for public health. The Mo-99, which results from U-235 fission allows following radioactive decay (66h $T_{1/2}$ period) the production of Technetium-99m (6h $T_{1/2}$ period), which is essential for diagnostic nuclear medicine. The current number of medical procedures worldwide that use Tc-99m is approximately 30 millions per year, of which 7 millions are in Europe.

At the European level, the production is primarily from the HFR reactor - NRG Petten, 45MWth (the Netherlands, 73% of European production [EP]), the BR2 - SCK-CEN Mol, 100MWth (Belgium, 15% EP) and OSIRIS - CEA Saclay, 70MWth (France, 12% EP). In Canada, the NRU reactor of AECL of 135MWth is a major world producer. In Australia, the OPAL reactor of ANSTO (20MWth, started into service in 2006) and in South Africa at Pelindaba, the SAFARI-1 reactor of NECSA (20MWth) both also produce Mo-99.

The current status is that most of the Mo-99 producing reactors are old (the majority started operation service in the sixties) and will soon reach the end of their operational lifetime. They will be taken out of operation in the coming few years and need to be replaced by new facilities in order to assure the continuity of radioisotope production in the next decades.

Within Europe, various projects have been started to replace the existing facilities:

In Netherlands, the Pallas Project in Petten will have a high level of production of radioisotopes.

From 2014 in France, the Jules Horowitz Reactor (JHR) [1], which is currently under construction, will allow the production of Mo-99 and will take the place of the OSIRIS reactor which will be stopped at that time.

The Molybdenum 99 production objective for the JHR reactor is to be able to guarantee 25% of the European base needs, with the possibility to increase the production up to 50% European needs if necessary.

Other new Mo-99 production projects are being studied, for example the FRMII reactor in Munich (Germany) [2].

The aim of this paper is to present the state of the investigation and planning carried out so far for the Mo-99 production facilities of the Jules Horowitz reactor.

This work was started in 2009 in cooperation with a team from NRG in Petten (HFR reactor) and CEA in Saclay (OSIRIS reactor).

The purpose of this work is to propose a reference architecture for the so-called MOLY circuits for JHR.

2. The JHR facility:

The Jules Horowitz Reactor (JHR) is a Material Testing Reactor (MTR). Its maximum thermal power is planned at 100MW and its design allows a large experimental capability (up to 25 experiments at the same time).

The neutron characteristics will make it possible to perform irradiations in the core which will provide high damage rates (up to 15dpa/year) in materials samples, while also carrying out in the reflector zone irradiations on fuel rods (cooking type, power ramp tests, etc) up to $500\text{W}\cdot\text{cm}^{-1}$. The primary circuit which cools the fuel elements in the core will be lightly pressurized (12 bars).

The reflector blocks, which will be made of Beryllium, will be cooled by a forced water circulation connected to the reactor pool (downstream flow).

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The JHR will be equipped with support systems that will allow the preparation and control of the targets for Mo-99 production before and after the irradiations phases.

Non destructive examination systems (both under water and in air) are foreseen in the pools and in the hot cells of the JHR building.

The locations for Mo-99 type radioisotopes production will be located in the Beryllium reflectors. After irradiation (approximately 6 days), forced cooling of the targets will be continued for some hours and then the targets will be prepared in a specific hot cell (called the ECR) and afterwards shipped by lead casks towards the Mo-99 producing facilities located at the present time in Belgium or in the Netherlands.

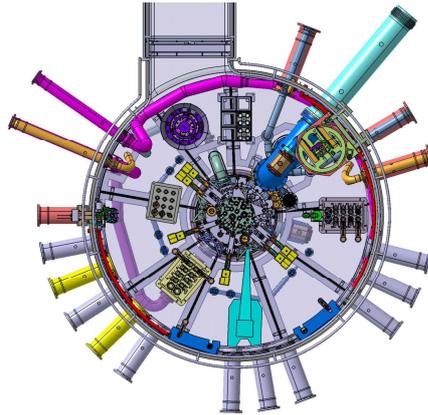


Fig.1: Top view of the JHR pool

Currently, the construction of the JHR is in progress. The reactor start-up is foreseen in 2014.



Fig.2: View on the JHR site (March 2010).

3. The JHR MOLY radioisotope production system :

To match the important Mo-99 radioisotopes production needs, the study carried out aimed at proposing a production facility in the JHR with a maximum capacity of 1000 targets/year that took into account different operating modes of the reactor (e.g. 70 MWth and 100 MWth).

The study was based on feedback received from the OSIRIS reactor located in Saclay (France) and also from HFR in Petten (Netherlands) to allow integration of the important experience gained at these facilities relating to target irradiations of the MOLY type.

- **3.1 Input data reminder :**

The production systems for MOLY targets will be placed in the JHR reflector in fixed positions and will also allow movable positions using displacements systems (called SAD). They will be localised in a sector of the Be-reflector that allows easy operation and will be positioned near the Nuclear Auxiliary Building (NAB) where the targets will be placed in the transport containers before being transported to the customers.

The systems (especially the movable positions) will offer a great deal of flexibility in radioisotope production. This flexibility also permits the possibility to modify the device configuration on displacement systems from a “radioisotope production” configuration to a “standard fuel rod irradiation” configuration.

The system will be designed according the ESPN regulatory rules and safety requirements (in terms of barriers).

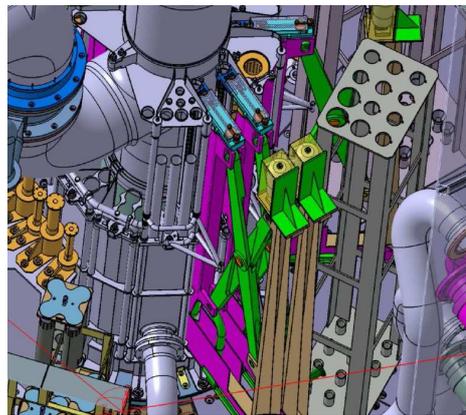


Fig.3: View on JHR MOLY sector.

- **3.2 OSIRIS reactor feedback :**

The OSIRIS reactor is a 70MWth MTR type. The core of the reactor can house up to 16 experimental devices in 4 positions where the fast neutron flux ($E > 1 \text{ MeV}$) ranges from 1 to $2.10^{14} \text{ n.cm}^{-2} \cdot \text{s}^{-1}$. Water boxes are also provided outside the core, on 3 sides of the vessel, making it possible to install up to 27 experimental devices on the first periphery, where the maximum fast neutron flux is 10 times weaker than in the core, and many others on the second and third peripheries. The maximum thermal flux in the middle of the side on the first periphery is close to $3.10^{14} \text{ n.cm}^{-2} \cdot \text{s}^{-1}$.

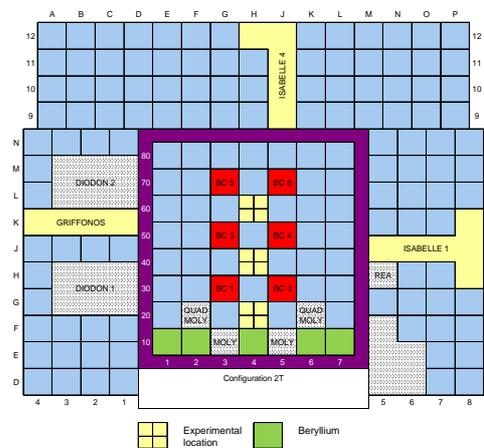


Fig.4: Experimental and MOLY locations in OSIRIS core

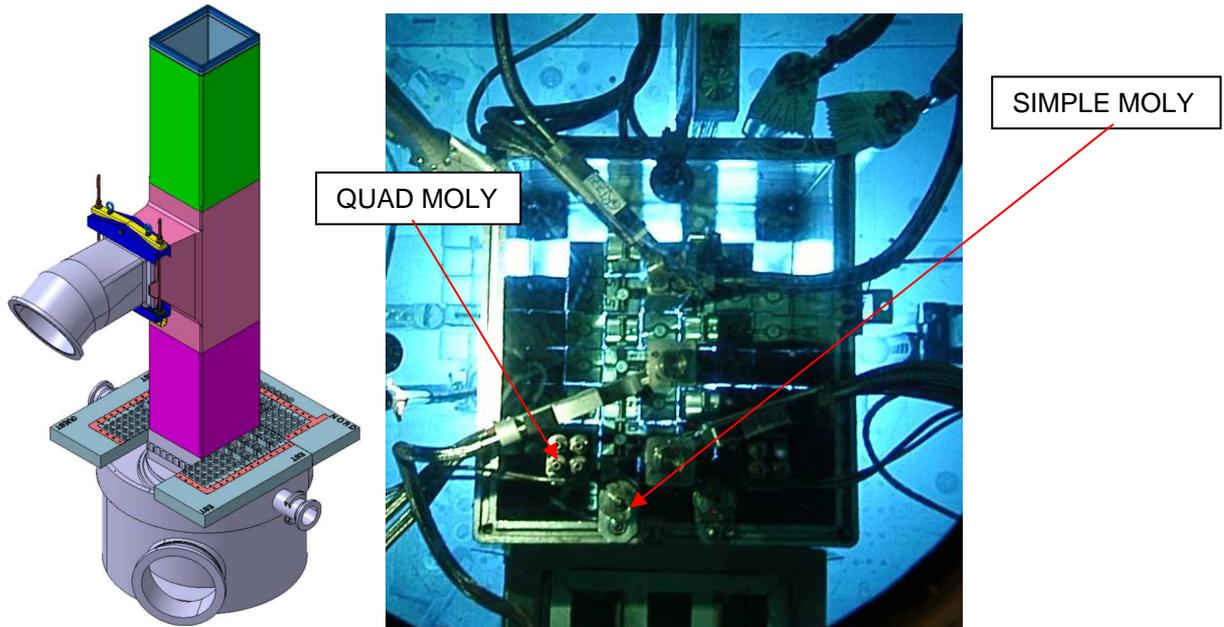


Fig.5: View of the OSIRIS tank and of the MOLY devices in the core

The OSIRIS reactor allows also the irradiation of MOLY targets. These targets are laid out in simple devices or in multiple devices of Quad MOLY type. In the core, there are 2 Quad MOLY and several MOLY locations available (these last are in the Beryllium reflector) .

The targets are placed by 3 on a target holder before being plugged into the devices.

Each device is placed in a cell of the core. Water of the primary circuit, in upward flow ($5500 \text{ m}^3/\text{h}$, 7.5 m/s), cools the fuel elements of the core, the in-core experiment and also the MOLY targets.

The advantage of this configuration is to have no specific coolant circuit for the MOLY (beneficial in terms of infrastructure, operation and maintenance). It also makes it possible to have a direct access with the targets during operation phases which facilitate the loading and unloading operations.

The configuration of the cooling circuit taking into account a chimney allowing to cool the targets in natural convection during unloading in slow speed with polar crane (outside neutrons flux).

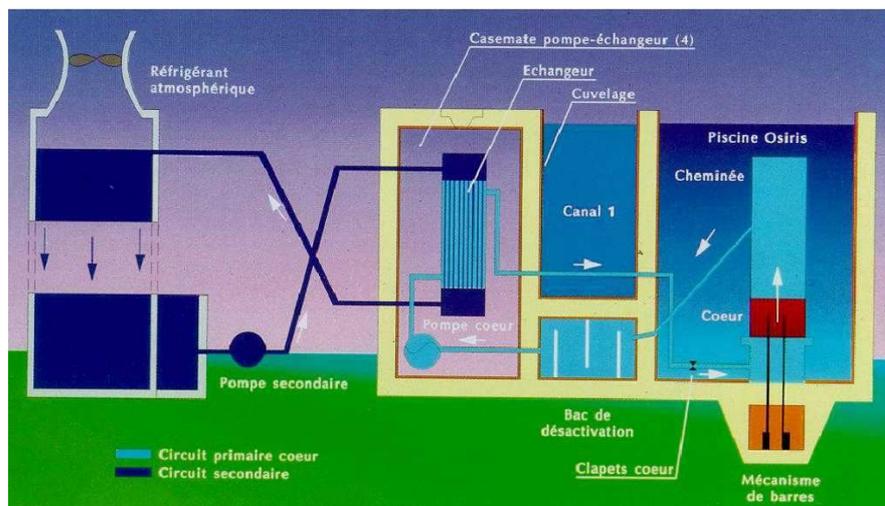


Fig.6: OSIRIS reactor: cooling circuit representation

After the irradiation, the targets are cooled in the pool during almost 30 minutes and then transferred in channel to be put in a basket. After time allowed for cooling, this basket of 6 targets is inserted in hot cell.

OSIRIS is equipped with 2 hot cells for sample preparation and MOLY loading in transport casks (up to 4 casks a day transported by 2 lorries). Each transportation cask contains 3 targets. The maximum activity allowed to be transported by cask is 2100 TBq. It is also possible to load casks under water.

In 2010, OSIRIS schedule of operation was adapted due to HFR and NRU outages; a very high number of targets have already been irradiated in 6 months, equivalent to yearly production during normal periods.

Quad MOLY device is used in OSIRIS reactor since 1989 and Simple MOLY device since 1981. Neutronic calculations are done before each OSIRIS operating cycle in order to establish fuel positions with different burn-up to avoid hot spots with respect to maximal linear power for thermal-hydraulic criteria. Sometimes the maximum power authorized in MOLY targets can limit the reactor power depending on different control rods positions.

The targets are tubular and highly enriched with ^{235}U isotope. The clad is a primary barrier, which means very strict controls during manufacturing.

The irradiation device is a water box, with 4 channels, locked at the bottom and on the core's superior grid. Each target-holder, with spacers, is introduced inside one channel with an irradiation pole connected and a handling mast. The handling mast is removed as soon as the target-holder is placed in the core. The irradiation pole includes a ballast to avoid fly off during irradiation due to the ascending flow in the core. In quad MOLY water box and simple MOLY sleeve, holes are drilled to evacuate cooling water in outlet primary pipe. The MOLY device doesn't contain any instrumentation.

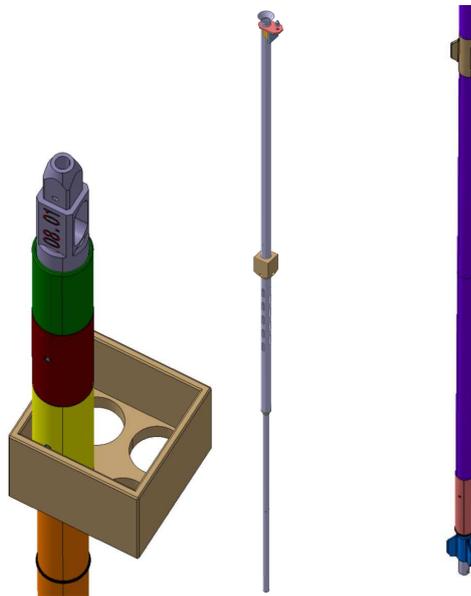


Fig.7: MOLY device description

During targets irradiation, clad tightness is monitored by CFD (Clad Failure Detection) system of the reactors' primary circuit. In case of clad failure, first anomaly threshold overshoot, filtration circuit is put in operation. Then in case of increasing values, emergency shutdown is automatically actuated. A mapping is done in order to identify which fuel element or MOLY device is concerned and unload it.

• **3.3 HFR reactor feedback:**

The HFR is a 45MWth reactor tank in pool type. The HFR is used for both material and fuel testing and for the production of radioisotopes (amongst others Mo-99). The irradiation of MOLY target is performed for both Covidien (plate-type targets) and for IRE (tube-type targets).

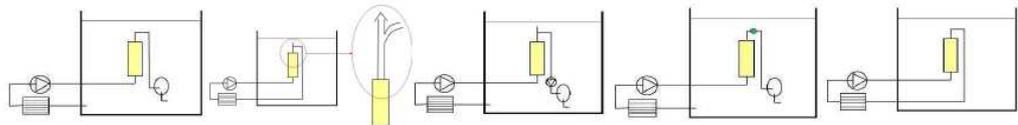
These targets are contained in a range of irradiation devices located in the core zone and in the reflector zone. The use of both plates and tube type targets and the use of in-core and out-of-core positions means that in total four different molybdenum irradiation devices are being used. The out-of-core positions have the advantage that the loading and unloading of the targets is relatively simple and that simpler irradiation devices can be used. The in-core positions have the advantage that the neutron flux is higher than the positions out-of-core. All four irradiation devices are constructed in such a manner that the targets can be loaded and unloaded from the HFR during full power operation.

The coolant circuits of the targets can be pressurized in the irradiation zone of the targets, thereby increasing the safety margin towards the Critical Heat Flux (CHF).

4. Research of a concept for MOLY circuits in JHR:

The first phase of the study was to propose different types of possible concepts for the new JHR MOLY circuits with a analysis based on different criteria, such as safety, design complexity, cooling performances, licensing regulations, Fission Product (FP) gas release management, etc.

The table below summarises the elements of this analysis:



Criteria / configurations	1	2	3	4	5
Exploitation (un) loading	Difficulty Lev.1	Difficulty Lev.0	Difficulty Lev.0	Difficulty Lev.2	Difficulty Lev.2
Safety	LOCA consequence studies Lev.3	LOCA consequence studies Lev.1	LOCA consequence studies Lev.1	LOCA consequence studies Lev.2	LOCA consequence studies Lev.1
FP gas trapping	FP gas trapping containment	FP gas trapping in the circuit	FP gas trapping containment	FP gas trapping containment	FP gas trapping in the circuit
Realisation	Difficulty, Level 1	Difficulty, Level 2	Difficulty, Level 3	Difficulty, Level 2	Difficulty, Level 2
Design	Difficulty, Level 1	Difficulty, Level 0	Difficulty, Level 3	Difficulty, Level 1	Difficulty, Level 2
CFD detection	Difficult (CFD in the pool)	CFD on REM circuit	Difficult (CFD in the pool)	Difficult (CFD in the pool)	CFD on REM circuit
¹⁶ N, FP releases problematic	Detection difficult Need circuit adaptation	Managed by design	Detection difficult Need circuit adaptation	Detection difficult Need circuit adaptation	Managed by design
Regulation	ESPN	ESPN	ESPN	ESPN	ESPN
Cooling performances	Low - MOLFI water comes from the pool	High – water comes from MOLFI circuit	Low - MOLFI water comes from the pool	Low - MOLFI water comes from the pool	High – water comes from MOLFI circuit

Fig.8: Comparative analyse of different concepts for JHR MOLY circuit

Legend: level 0 (simple) - level 1 (medium) - level 2 (complex) - level 3 (very complex)

As a conclusion of this analysis, a choice of the most favourable concept has been made.

The main characteristics are mentioned hereafter:

- The cooling water circuit of the JHR MOLY targets will correspond to a nearly closed cooling circuit being able to function both at a JHR power of 70 and 100MWth. This configuration represents the best compromise taking into account the various criteria.

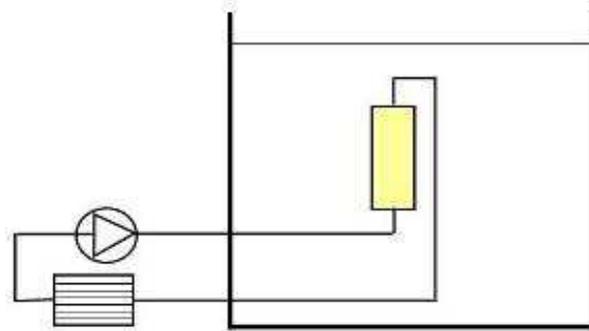


Fig.9: Concept proposal for MOLY fluid circuit.

Note: The figure indicates the main components of the circuit as pump, heat exchanger, irradiation device in reactor pool.

- The cooling circuit for MOLY production will be a circuit independent of the reflector coolant circuit (called REP). It will be able, according to the working modes (i.e 70 or 100MWth) to be pressurized until approximately 10 bars in order to increase the cooling capacity.
- The zones in the reflector will concern the fixed sectors and sectors located on displacement systems. This area is closest to the channel transfer of the targets towards the Nuclear auxiliary building (where are located the hot cells used for target conditioning after irradiation).

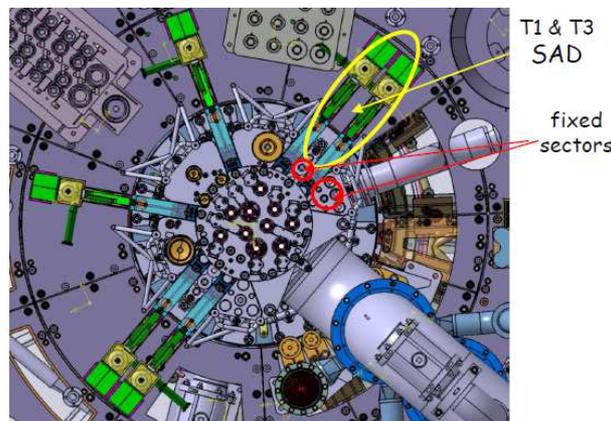


Fig.10: View of the MOLY sector in the JHR core.

- The displacement system will be able to accommodate devices for the production of radioisotopes but could be reconfigured in order to carry out standard irradiations of fuel rods. Each irradiation MOLY device can embark several targets (up to 12 targets / device).
- The circulation of the coolant water will be downward over the targets. The fluid velocity around the targets will be approximately 7 to 8 $\text{m}\cdot\text{s}^{-1}$. The devices will include in the upper part a zone of

forced convection cooling (“chimney” arrangement) to cool the targets after irradiation and before unloading from the device.

- Any fission gas releases (in accident conditions) will be managed by placing a sealed removable lid on the top of the device. The detection of any activity released in the circuit will be measured by a CFD (cladding failure detector) located in the coolant circuit in the cubicle (out-of-pile part of the MOLY circuit).
- The hydraulic components (circulating pumps, heat exchangers, CFD) will be placed in a bunker located close to the MOLY production sector.

The devices both in the fixed and movable sectors will be very similar in term of design in order to facilitate easy operation (i.e loading and unloading of the targets).

5. MOLY circuit architecture proposal:

Below is the currently proposed description of the MOLY circuit:

The flow circuit will allow the cooling of the MOLY devices in both the fixed sector and in the reflectors.

The coolant water circulation is ensured by pumps (for normal operations and one back up for safety) which will work in parallel under normal conditions.

In the event of failure of the main pump, the safety pump will ensure the cooling of the targets with a flow defined by the safety studies. The instrumentation on each line will be composed of pressure sensors and flow meters. CFD (cladding failures detectors) will be fitted in the circuit in order to detect any cladding rupture. In this event occurred, the reactor will be stopped and the cooling of the targets will continue using forced convection.

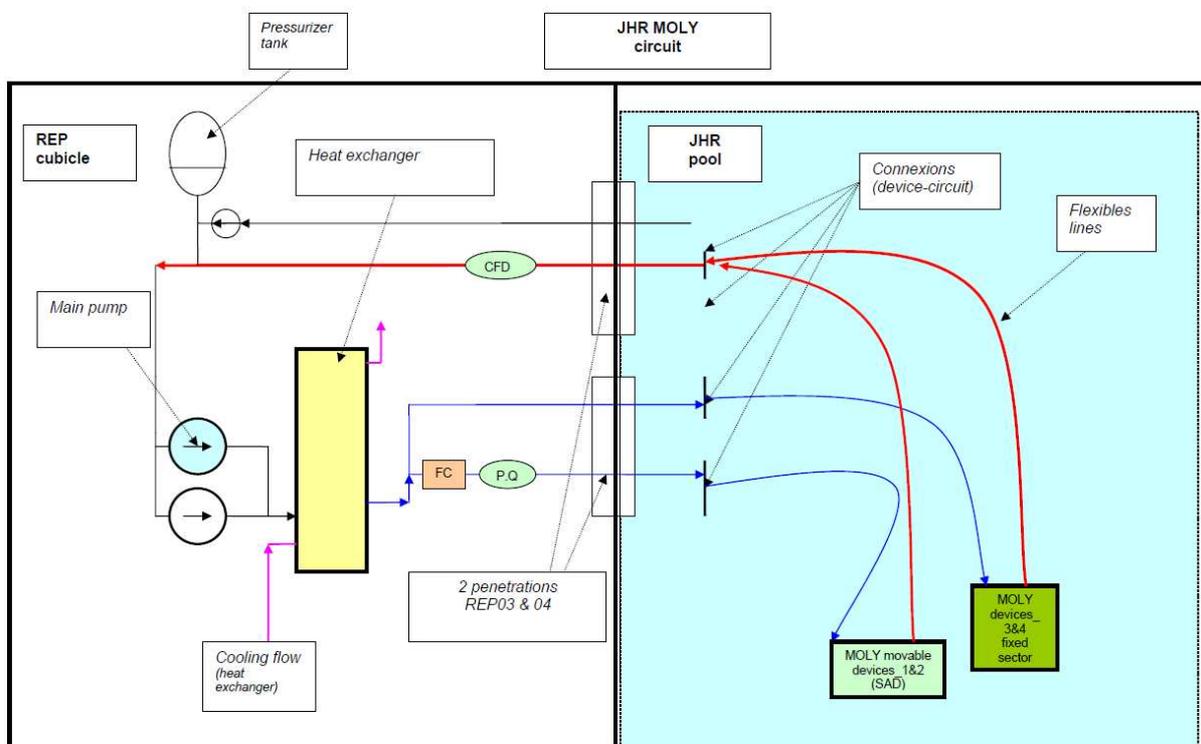


Fig.11: Simplified representation of MOLY coolant circuit

6. Neutron data:

The sectors chosen for the MOLY devices locations offer the following performances in terms of neutron characteristics:

Facility	Location	Thermal neutron flux (perturbed) $10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$	Note
OSIRIS	Sector 20	1,2 to 1,8	70 MWth
JHR	Fixed and movable sectors	Target value (minimum) 1,5	Objective at 70 MWth

7. Operation scenario proposal:

- On their arrival in the JHR facility, the new targets are stored before being used,
- The targets are placed under water in the target holders which are able to contain up to 12 targets for the devices of the Quad MOLY type,
- The target-holders are inserted in a irradiation device located in the reflector using handling tools,
- The target holder is locked in the device for the irradiation phase which is approximately 6 days duration,
- For unloading, the target-holders continue to be locked to the device, which is moved to an intermediate position with “forced cooling flow”. This phase will last a few hours and will be sufficient to reduce the residual heat power of the targets to a level acceptable for transfer,
- The targets are then unloaded from the targets holders and are transferred under water to the hot cells. The storage duration of the target for cooling in natural convection is approximately 5 hours,
- The targets are introduced in hot cells and then are transferred in a lead flask accosted with the ECR hot cell,
- The lead flasks are evacuated and are shipped to the Mo-99 production facilities.

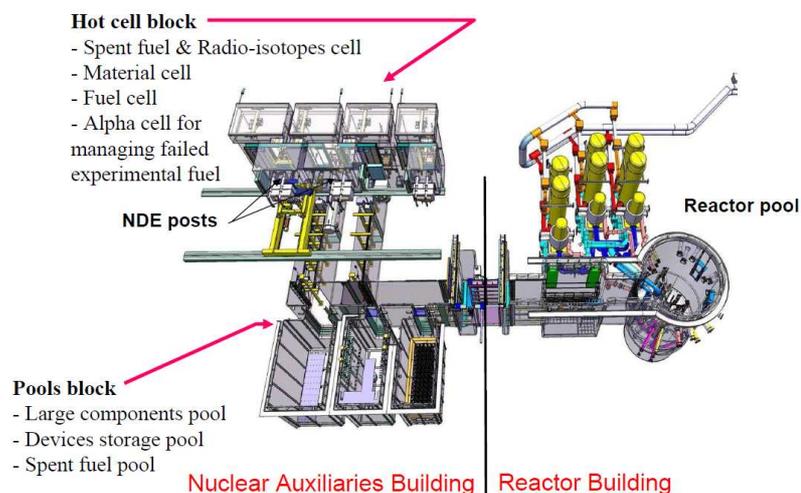


Fig.12: View of the under water transfer channel in the Nuclear Auxiliary Building (NAB)

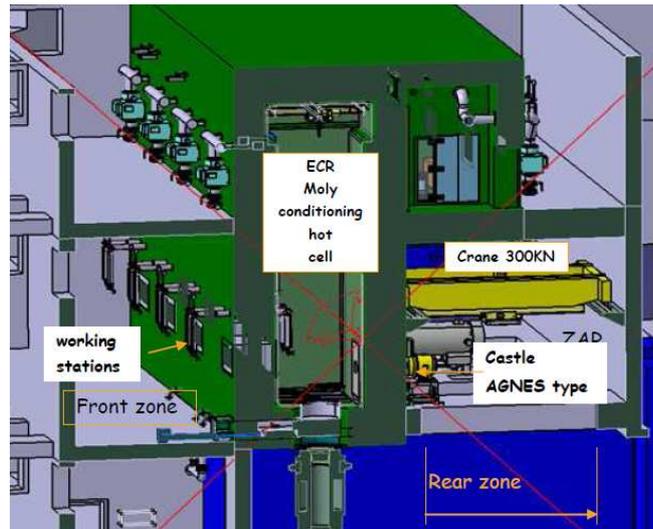


Fig.13: Side view on the Hot cell for MOLY targets conditioning

8. Conclusion and next steps:

After the preliminary phase of analysis and research of concepts, reference architecture for JHR MOLY circuit has been proposed.

The next phase will concentrate on the conceptual design of the MOLY circuit, the irradiation devices and targets holders (3D CATIA design studies, thermo-mechanical dimensioning studies following RCC-MX rules, ESP(N) regulation verification, etc.).

In parallel, a preliminary analysis of safety will be performed to evaluate the consequences following a theoretical rupture of the MOLY coolant circuit (LOCA type).

An analysis will be also performed in order to determine the necessary level of redundancy of components (e.g. number of pumps, heat exchangers) in order to ensure the best availability of the MOLY circuit for production while costs and benefits.

9. References:

[1] Jules Horowitz Reactor - Experimental capabilities

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