Transfer of R&D accomplishments between ISOL facilities

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Physics and Technology for the Next Generation of Radioactive Ion Beam Facilities: EURISOL

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1. Introduction

Since the discovery of artificial radioactivity in 1935, nuclear scientists have developed tools to study nuclei far from stability. A major breakthrough came in the eighties when the first high energy radioactive beams were produced at Berkeley, leading to the discovery of neutron halos. The field of nuclear structure received a new impetus, and the major accelerator facilities worldwide rivaled in ingenuity to produce more intense, purer and higher resolution rare isotope beams, leading to our much improved knowledge and understanding of the general evolution of nuclear properties throughout the nuclear chart. However, today, further progress is hampered by the weak beam intensities of current installations which correlate with the difficulty to reach the confines of nuclear binding where new phenomena are predicted, and where the r-process path for nuclear synthesis is expected to be located. The advancement of Radioactive Ion Beam (RIB) science calls for the development of so-called next-generation facilities, which will provide beam intensities several (2-4) orders of magnitude higher than presently available, and provide us with many isotopes currently inaccessible. In particular in Europe NuPECC, the European Coordination Committee for Nuclear Physics, recommends building the next generation ISOL installation EURISOL as the highest long term priority for low energy nuclear physics. The physics case and technological solutions for EURISOL were laid out during the EURISOL Design Study, which brought together 20 laboratories representing 14 European countries and was partially funded by the European Commission during the 6th framework program. CERN was a major participant in this study and was recognized as one of the possible sites for the future facility.

2. Physics case

a. Nuclear Structure

The first major result stemming from the inception of radioactive ion beams was the discovery of neutron-halo nuclei, characterized by dilute neutron matter surrounding a tightly bound core. The large extension of the neutron wave function makes the ^{11}Li nucleus appear the size of ^{208}Pb. Heavier halo nuclei are also predicted by theory, such as ^{14}Be, ^{19}C, ^{23}O but have been scantly studied up to now due to low beam intensities. For heavier neutron rich nuclei the halo structure is expected to give way to a more compact neutron skin. These extended neutron distributions have major consequences for nuclear structure and excited states,. It has been observed that magic numbers, once thought to be immutable, are in fact quite fragile when large neutron to proton ratios are reached. For example, detailed studies of Oxygen and Magnesium isotopes have shown that the magic neutron number 20 is replaced far from stability by N=16. Reducing the number of protons for a given neutron number will modify the n-p interaction and hence the relative position of the single particle levels. The more diffuse density distribution due to a neutron skin should reduce the intensity of the surface peaked spin orbit interaction and therefore the shell gaps in the heavier nuclei which are known to be spin orbit driven. New types of excitations also appear, in particular oscillations of weakly bound neutrons against the tightly bound core called pygmy resonances due to their low energy and cross sections in contrast to the well-known giant resonances.

The understanding of the structure in a region of the nuclear chart requires a multifaceted approach over long isotopic chains including the measurement of ground state and excited state properties. The experimental approaches can be roughly classified in two categories: stopped beam experiments for the measurement of ground and isomeric state properties and radioactive decay and

reaction experiments where states are excited through Coulomb or nuclear interactions. EURISOL will bring extraordinary advances to both types of experiments through unmatched intensities and a uniquely broad energy range spanning from keV/nucleon up to above 100 MeV/nucleon. One can expect for example to measure the mass and charge radius of ^{78}Ni as well as magnetic moments in this region. Combined with single particle energies obtained through the analysis of transfer reactions, these data will allow the magic character of Z = 28 and N=50 to be probed far from stability along the r-process path. In the lighter region the relics of doubly magic nuclei under the form of resonances such as ^{28}O will become accessible. Figure 1 shows the chart of the nuclides along with the main physics subjects for EURISOL.

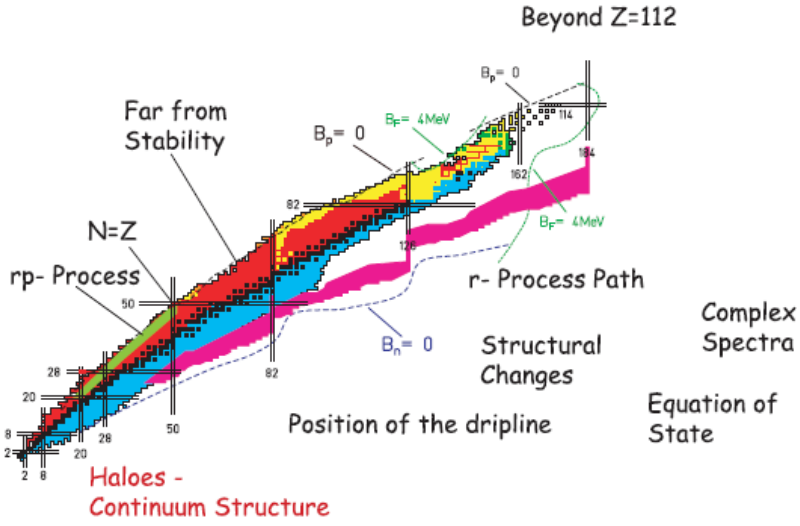


Figure 1 : Chart of the nuclides and main physics reach for EURISOL

EURISOL will also provide new insight into the existence and the structure of the heaviest of elements. The decay properties of their very neutron-rich isotopes will be investigated. The major goals of the experimental programme will be to push towards the predicted closed neutron shell gap at N=184 and to obtain systematic data on nuclear decay modes, half-lives and masses at the upper extreme of the Segré chart. Isotope shift measurements and hyperfine spectroscopy will be performed on various isotopic chains. The experiments will reveal the location of atomic levels and allow the determination of nuclear ground-state properties such as deformation and spin.

These new manifestations of nuclear structure are a challenge to theory. The results from EURISOL experiments will furnish major constraints for modern nuclear theories and enhance their predictive power for the properties of nuclei at the confines of the nuclear chart which may never be reached experimentally.

b. Nuclear Astrophysics

The bulk of the nuclear species heavier than iron are produced in rapid capture processes of neutrons or protons followed by radioactive decay, the so-called r-process, which is thought to take place during explosive supernova events. EURISOL will produce copious amounts of the radioactive nuclei involved in the r-process, providing the necessary values of nuclear masses, lifetimes and capture cross sections for input into the astrophysical models. The magic numbers far from stability greatly influence the r-process path and the final isotopic abundances – our expanded

knowledge of exotic nuclei will produce the missing links for astrophysicists to fully understand the nucleo-synthesis of heavy nuclei.

Neutron stars are the remnants of core-collapse supernovae and are the most compact stellar objects after black holes. Many of their properties such as internal composition or temperature cannot be directly linked to observations but much of the missing information can be obtained from the study of unstable atomic nuclei which will be available at EURISOL. Of particular importance for the modelling of these stellar objects is the knowledge of collective excitations such as pygmy resonances, the understanding of nucleon pairing in exotic nuclei and a foray into nuclear thermodynamics through the evolution of the nuclear equation of state with increasing neutron to proton ratio. The latter exploration requires high energy exotic nuclei in order to excite their stiff giant monopole resonance and to induce their multi-fragmentation through violent nuclear collisions.

c. Fundamental Interactions

The study of nuclear decay modes has played a crucial role in determining the basic structure of fundamental interactions. For example, precision measurements of beta-decay have provided many experimental foundations for the standard model. The measurement of super-allowed $0^+>0^+$ transitions leads to the determination of the semi-leptonic weak coupling constant G_v and consequently of the V_{ud} element of the Cabbibo, Kobayashi, Maskawa (CKM) quark mixing matrix. EURISOL will allow such studies for N=Z nuclei up to ^{98}In much heavier than accessible today. These measurements will also lead to new insight into isospin mixing in nuclear states.

Correlation measurements in nuclear beta decay are a very sensitive tool to investigate the presence of exotic (i.e. scalar or tensor type) weak interaction components, search for right-handed charged weak currents and search for new sources of time reversal violation. This type of precision physics can lead us beyond the standard model. Such measurements will greatly benefit from the large intensities delivered by EURISOL, but will also necessitate the development of new types of ion and atom traps.

The beta-beam concept for the production of high intensity electron (anti-)neutrino beams for oscillation physics was first proposed by Piero Zucchelli at CERN in 2002. The EURISOL beta-beam facility is adapted for a base-line of some 130 km corresponding to the distance between CERN and the laboratory installed in the Frejus tunnel. The beta-beam facility is designed to use the existing CERN injector chain.. The chosen energy and baseline makes the facility an excellent tool for the study of CP violation for a large value of θ_{13} (>0.001), which seems now to be observed in the most recent reactor experiments.

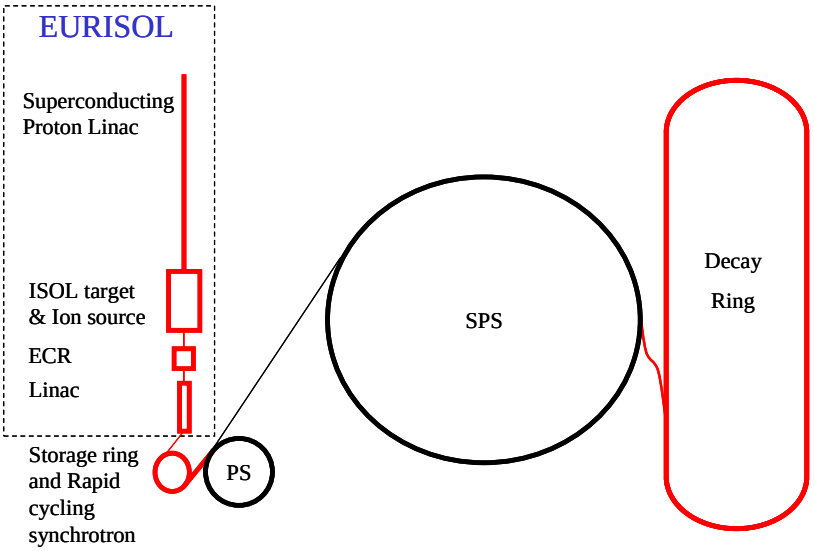


Figure 2: Schematic view of the beta-beam facility

3. Technical solutions

The planned EURISOL facility will use a large Continuous Wave (CW) superconducting linear accelerator (the "driver" accelerator) to accelerate H^- ions to energies of 1 GeV. The option of using a pulsed beam at 50 Hz with a minimum pulse length of 1 ms has been kept open to enable possible sharing of the driver with other physics users. This beam of particles will deliver a power of up to 4 megawatts to one target station, and through a newly developed magnetic beam splitting system some 100 kilowatts to three smaller target stations in parallel. To achieve these ambitious goals the low energy section of the linac will use a RFQ and Half Wave Resonator linac. The medium-low energy section is presently designed using the newly developed triple spoke cavities from IPNO in Paris while the medium energy section uses five cell elliptical cavities developed at CEA in Paris. Finally, the high-energy section will also use the five gap elliptical cavities adapted for the higher energy. The design has been optimized for beam quality and cost.

In an ISOL converter system, the neutrons are generated by high-energy protons impacting on a high Z material (so called spallation n-source). The radioisotopes are the fission products of fissile target material positioned close to the neutron source. In order to cope with the 2.3 MW power deposited in the spallation target, out of the 4 MW EURISOL proton beam, the converter has to be made of liquid metal. Two options based on axial or radial molten metal flow directions were investigated. Conceptually, several targets filled with ^{235}U or other actinides are inserted, through a channel created in the shielding, close to the neutron source at the position of maximum neutron flux. The neutron flux is thermalized in order to optimize ^{235}U fission while for other fissionable target materials, like ^{238}U or ^{232}Th, a hard neutron spectrum is required. Up to six targets can be positioned simultaneously, linked to single ionization ion sources (laser, plasma, ECR).

Up to three direct targets, in which the target material is directly exposed to the proton beam will also be available simultaneously. The main challenge set by the EURISOL beam power, is that the evacuation of the energy deposited by the 1 GeV protons through ionization in the target material, while the target materials (some of which are low density and open structure materials or are in the form of oxides with thermal insulating properties) are kept at the highest possible temperature to minimize the diffusion time of the radioisotopes.

The beam from the target stations has to be prepared for experiments with the merging, cooling, mass-separation and charge state multiplication of the six beams from the multi MW fission targets representing the biggest challenge. Preliminary studies have been performed of merging using a so-called arc ECRIS source which has geometry suitable for injection of several beams into an ECR plasma from which a single beam later can be extracted. The transverse cooling will be done in a newly developed high intensity RFQ cooler which also permits pulsing of the beam for experiments needing a specific time structure. The mass separation will be done with a classical dipole systems consisting of up-to four independents dipoles which should be capable of isobaric mass separation providing that the radioactive beam can be transversely cooled to a sufficiently small beam emittance. The exotic ion species from this area will then be directed towards low-energy experimental areas, where they can be captured in magnetic "traps" so that there properties can be studied, or led into the post-accelerators. Prior to post acceleration, the necessary charge breeding will be done in either an ECR source or in a new high intensity CW EBIS source.

There will be at least one superconducting linear accelerator, up to 200 meters long, in which exotic ions will reach energies up to 150 MeV (million electron-volts) per nucleon. The high-energy linac has been optimized for ions with mass-to-charge ratio (A/Q) up to eight with a final top-energy of 150 MeV/u for $^{132}Sn^{25+}$ which has been selected as the reference beam. The linac consists of only three different cavity types: independently phased quarter wave resonators, half-wave Resonators, and spoke cavities. The post-accelerated beams will have sufficient energy to undergo secondary fragmentation, leading to neutron-rich nuclei further from stability than those produced by any facility existing or under construction today.

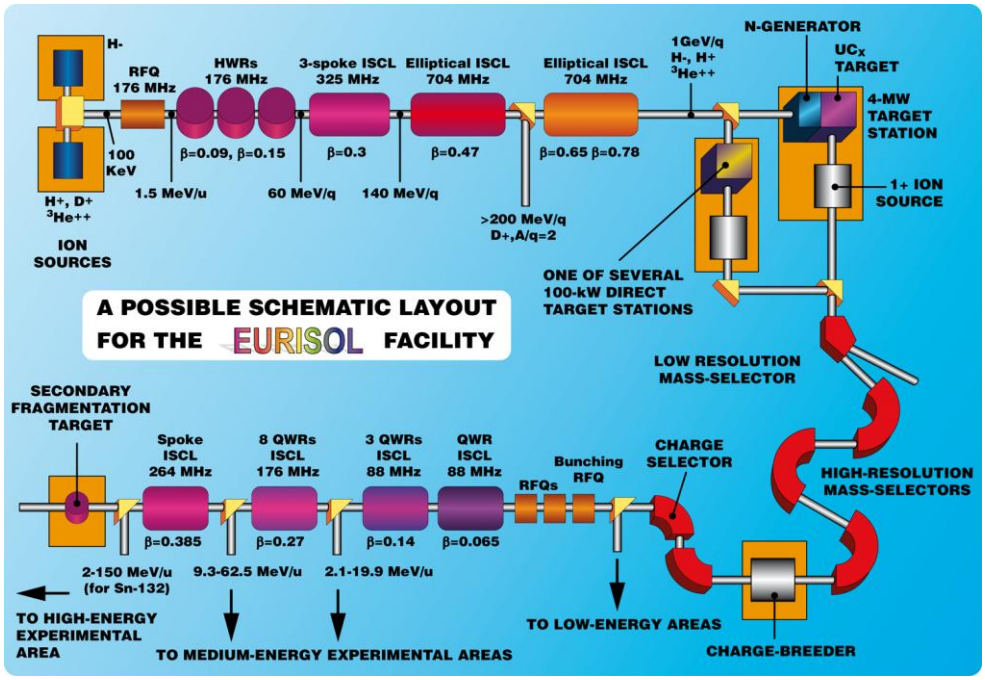


Figure 3: Schematic view of the EURISOL facility

4. Technological developments

Since the end of EURISOL-Design Study in 2009, a number of new large infrastructures that share common technical challenges in Europe and elsewhere are under design or have started their construction phase. The most relevant ones are Myrrha in SCK-CEN Mol, Belgium, ESS in Lund, Sweden, and ARIEL in TRIUMF, Canada. Myrrha is an ADS demonstrator. It will operate a 600 MeV, CW, 4 MW proton linac which will impact onto a liquid Pb/Bi eutectic target, part of the cooling system of the sub-critical core of a nuclear reactor. ESS in Lund is the European spallation neutron source that will operate a "long" pulsed proton Linac of 2.5 GeV, 14 Hz, 2.8 ms, 5 MW on average, intercepting a rotating tungsten target cooled with helium gas. Finally, ARIEL will be operating a 50 MeV, CW, 0.5 MW electron linac, aiming at producing 10^{14} fissions/s in a refractory actinide target to deliver neutron rich radioisotope beams, and will be complemented with a more traditional 500 MeV, CW, 100 kW, proton beam sent onto an ISOL target alike ISAC II presently in operation. This rapid overview should furthermore be completed with operational feedback obtained from SNS in Oakridge, the American neutron spallation source operating a pulsed 1 GeV, 1 MW proton linac onto a liquid mercury loop target.

In view of the elements obtained from these different high power facilities, and integrating recent evolutions in the way beams are delivered at ISOLDE at CERN, different key elements of the EURISOL facility needs to be specifically addressed in the coming 5 to 10 years:

a. Superconducting Linear Accelerator Technology

Advances in RFQ systems and superconducting accelerator cavities are necessary for the realization of the HIE-ISOLDE and SPIRAL2 projects and later for EURISOL, for both proton driver and heavy ion post-accelerator. At CERN activity driven by R&D towards HP-SPL for proton driver and HIE-ISOLDE for the post-accelerator and the low and intermediate energy of the driver. Strong synergy with ESS at Lund and ISOL@MYRRHA at CEN-SCK, Mol, Belgium.

b. Neutron converters

Converters which produce neutrons from initial charged particle beams are an essential component of high power ISOL facilities. Solid converters are in use at ISOLDE and being developed at GANIL for SPIRAL2.

Within the EURISOL Design study, the target complex was one of the key elements of the EURISOL facility, which was the subject of intense design work and preliminary prototyping. The 4 MW mercury converter was to be set up in a target station where it would be utilised in conjunction with a uranium target to create exotic isotopes by rapid fission. The converter was based on a circulating metal loop exposed to direct proton beam irradiation. First prototypes were developed and tested off-line, from which relevant experience was gained. At the present stage, several issues are yet to be addressed, such as the impact of beam irradiation parameters on the liquid metal loop operation. Likewise, the target development will necessitate further research in the field of heat exchange with liquid metal, irradiation, corrosion and fatigue testing of materials.

Achieving the design of the full-scale target for EURISOL will require several partial tests: first of sub-components, irradiation at lower power, through to instrumentation tests, and finally full-scale tests. In order to better ascertain the needs for testing a high-power target, various existing and projected facilities have to be examined and their characteristics evaluated. These issues will be partly addressed in the WP9 task of the TIARA project lead by CERN.

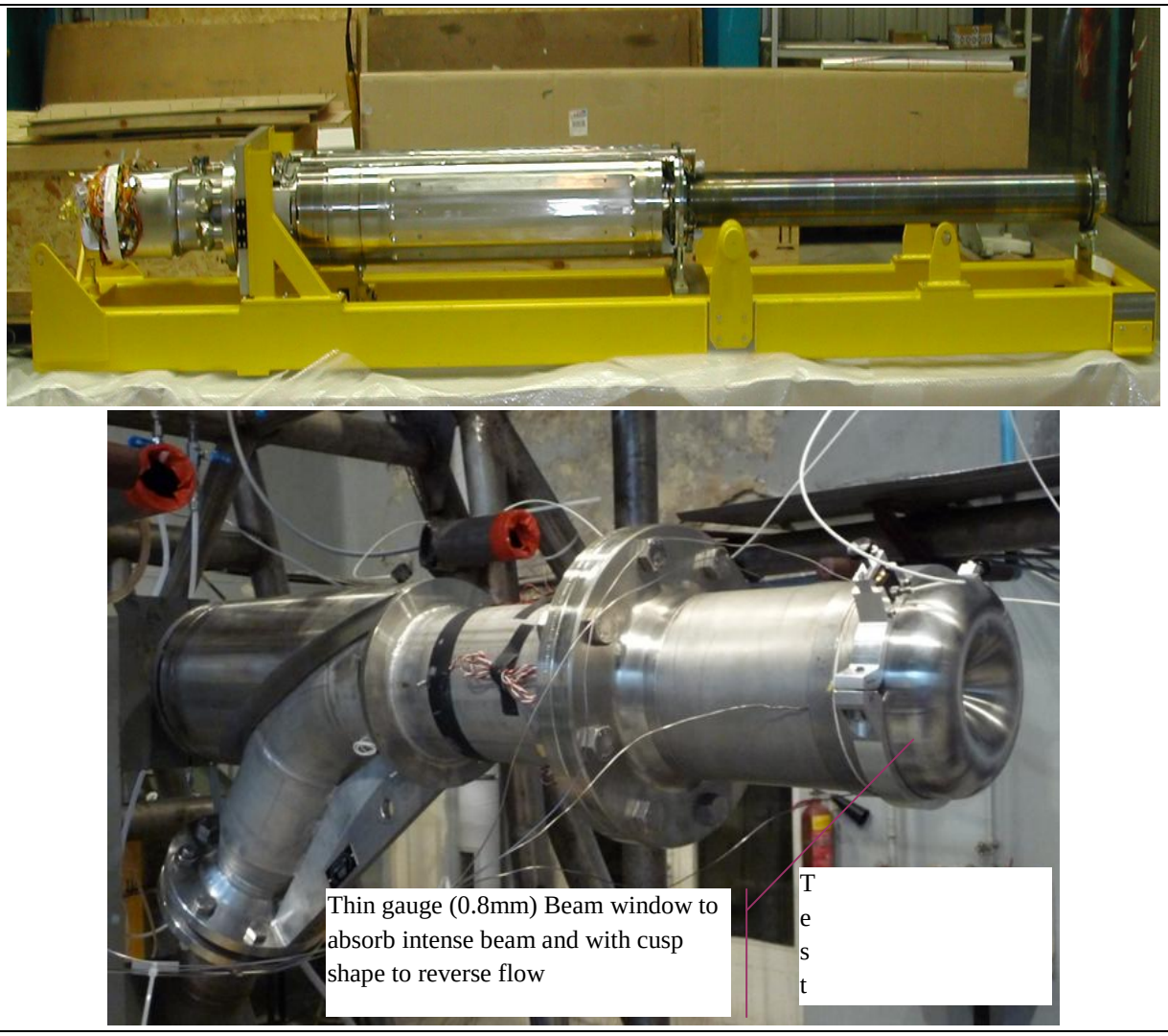
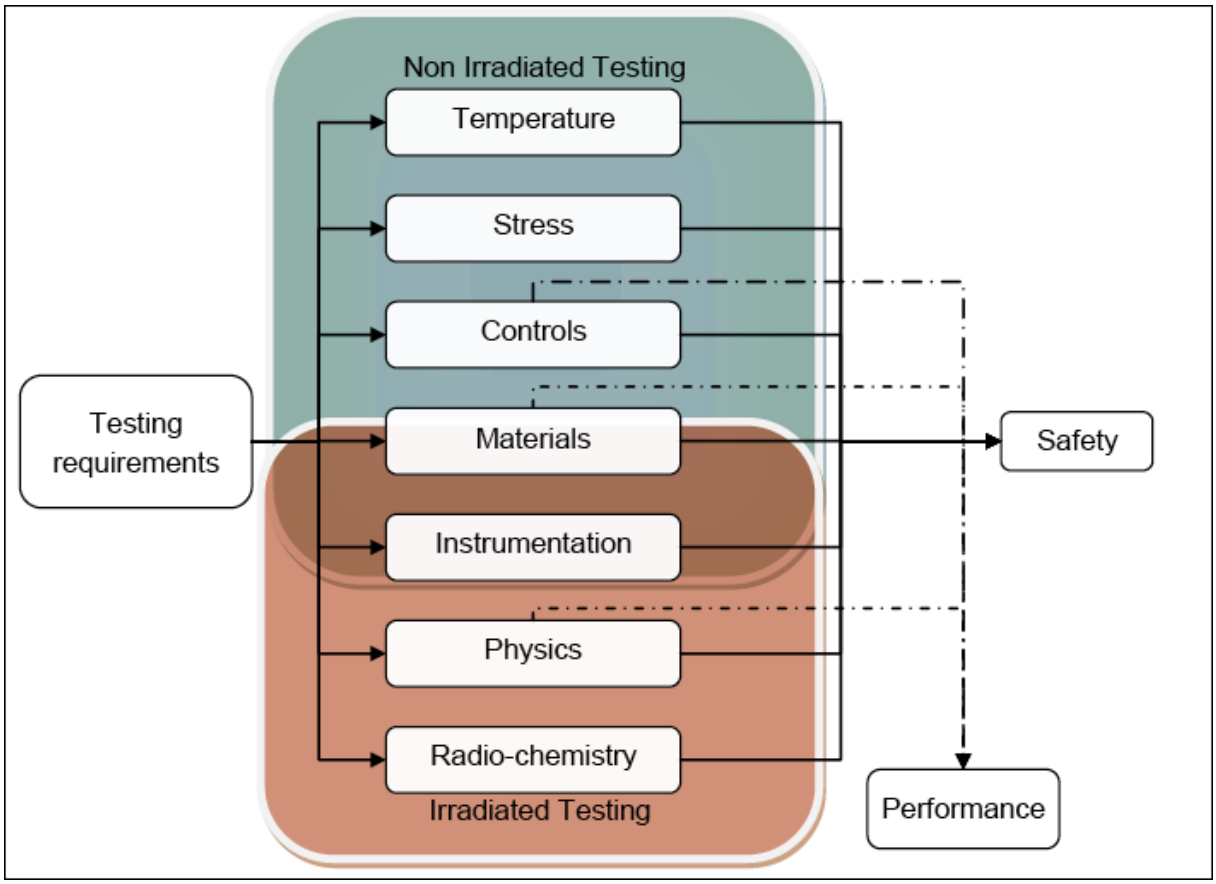


Figure 4: Liquid metal neutron source Megapie (top), Eurisol on test stand (bottom)

Testing liquid metal sources require setting up a liquid metal loop with all measurement devices required for measuring pressure, temperature flow and local velocities where needs. Prior experience gained in this respect indicates that selecting specific components for dedicated tests such as the beam window is a proven way forward. In a graded approach aimed at minimising risk, integral testing is required only once the component testing is successful.

In order to proceed in a cost-efficient manner, the different aspects of target testing have been broken down into separate testing requirements to avoid replicating large facilities or placing such high demands on the proposed facilities, that they would make the project unaffordable. Each requirement can thus be assigned to a specific facility. Where compatibilities exist, adaptations are

to be made; otherwise a dedicated facility would have to be built. By focusing on a particular testing requirement, it is however far more likely that an existing facility may accommodate the specific testing needs, failing which the purpose-built facility which would have to be built would focus on a narrower set of objectives and thus be cheaper.



Ideally both solid and liquid metal targets should be developed in parallel so as to promote dissemination of knowledge. Some of the facilities may then be shared between a solid target testing station and a liquid metal target testing station if both options are pursued.

c. Development of fission and other target materials

The improvement of radiation hardness and release properties of ISOL targets are essential to increase the beam intensities and scientific output of ISOL facilities. Vigorous related R&D, particularly concerning sub-micron structured target materials, are performed at ISOLDE. This has already been witnessed using solid targets with nanostructures, and should be translated to the most important class of Uranium carbide targets within the WP8 in FP7-ENSAR. If the gain of at least one order of magnitude is confirmed for the most exotic isotopes, this would pose the question as whether a MW-class primary beam power is still required to meet the EURISOL physics goals, with implications on cost, site selection decision and safety aspects.

In line with the precedent results, an important prototype tests is foreseen in 2014 on a liquid metal target loop equipped with a diffusion chamber. This should indicate if isotope release properties can be improved by fragmentation of the molten metal into thin droplets.

A final axis of development consists in the production of beams of refractory elements that were until then not accessible by the ISOL method. Such beams are available by fragmentation of the post-accelerated (non-refractory) secondary radioisotope beams within EURISOL-DS. The method consists in the release of the refractory elements through volatile molecule formation and subsequent break-up in the ion source cavity, either by laser light or by electron impact.

d. Improvement of Ion Source performances

Extracted as singly charged ions from the target-ion source units, the radioactive isotopes have to undergo a charge breeding process to an n+ state to match the limit in mass-to-charge-ratio of the post-accelerator. The study and development of charge breeding techniques plays a prominent role for optimizing the post-acceleration of intense and exotic beams that will be produced in EURISOL. High charge states, i.e. relatively low A/q-ratios, allow for compact ion accelerators and higher final beam energies, in particular in combination with superconducting LINAC structures. This post-acceleration scheme, also known as 1+ n+ scenario, presents several technical challenges because of the diversity of the produced isotopes in terms of mass (spanning the complete nuclear chart), lifetime (short lived, 1ms to stable), produced intensities (from a few up to 1E13 ions/s) and because of the combined rareness and short lifetimes of the most exotic isotopes.

Because of the challenges listed above, the charge breeding technique used has to be universal, rapid, efficient and needs to deliver sufficiently high charge states to allow the post-acceleration of ISOL-type ion beams produced by a EURISOL-like facility. Other parameters may additionally influence the choice of technique. The time between system failures, the time to repair and the maintenance requirements may also have to be considered in view of radioprotection issues, while different aspects of flexibility includes ease of charge state selection and continuous wave (CW) contra pulsed operation capabilities.

Up to now, mainly three charge-multiplication techniques are in use for the post-acceleration of radioactive beams. The first one is the stripping technique based on acceleration of low charged ions followed by subsequent stripping in a gas jet or thin foil. Although it is a very efficient method for the production of bare light ions, a lower efficiency is experienced for heavy ions for which the post-stripping charge-state distribution is wide, and multiple stripping stages have to be used. It also requires a costly pre-stripper section, with low frequency RF-structures for the extreme A/q-range, that accelerates the radioactive ions to the minimum energy needed for the stripping process. Multi-charge state acceleration is an attempt to improve the overall efficiency, at the cost of worsened longitudinal and transverse emittance properties. The stripping method, although the most rapid and robust one, might not be the best choice for EURISOL due to the drawbacks given above. The two other charge breeding techniques make use of either an Electron Beam Ion Source (EBIS) or an Electron Cyclotron Resonance Ion Source (ECRIS) as charge breeders. Apart from EURISOL, many facilities are presently investigating or developing EBIS/T and ECRIS charge breeders. Among them, SPIRAL2, SPES, the CARIBU project at ANL and TRIUMF/ISAC will use ECRIS charge breeders. On the other hand MSU is developing an EBIT charge breeder for FRIB based. In this context the two techniques of charge breeding are expected to evolve rapidly, and second generation EBIS and ECRIS charge breeders are already being built or tested. This section will therefore focus on EBIS and ECRIS charge breeding methods.

The EBIS charge breeding concept combines an EBIS with a preceding Penning trap or RFQ cooler for accumulation, bunching and cooling of the beam. It has been pioneered at REX-ISOLDE and there provided beams with masses ranging from 8Li to 224Ra, and with very different half-lives and chemical properties (alkali, metallic and noble gas ions), including fragments of molecular beams coming from ISOLDE [1]. The inherent properties of an EBIS facilitate breeding of ions to high charge states combined with low residual gas content in the extracted beam, however, space charge limitations, primarily set by the Penning trap, limits the injected current to around 1 nA. The machine is best operated in pulsed extraction mode, favoring the injection into pulsed LINACs.

In the ECRIS charge breeder, the ions are injected directly without prior preparation. The device has a very high current capability, exceeding 1 uA injected current. It is a less complex system which demonstrates a high reliability. The extracted CW beam is well adapted to cyclotrons and superconducting LINACs, although the device can be operated in pulsed extraction mode with some ms extraction time. Presently the performance is hampered by the large current of residual gas ions produced on top of the charge bred radioactive ions.

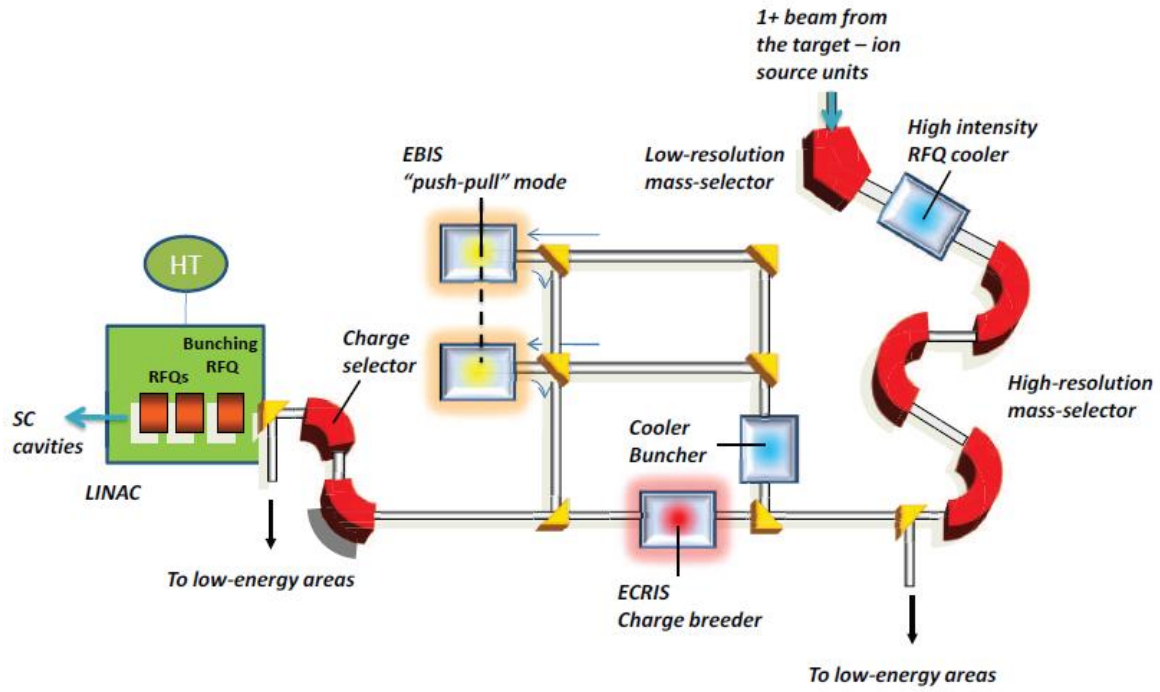


Figure 5: Detail of the EURISOL layout accommodating both charge breeders. Optionally the two EBIS will be run in "push-pull" mode for a pseudo-CW operation. Taken from [2].

Based on an evaluation of the two charge breeding methods performed within the EURISOL framework, a solution can be proposed that should satisfy the needs of a future facility by using both charge breeders types in parallel and their complementary features (see Fig. 5). In summary the abilities of both breeder systems are:

- Charge states yielding mass-to-charge ratios between A/q=2-3 and A/q=7 can be obtained for all elements in the chart of nuclides, with lowest A/q for EBIS.

- Efficiencies well above the percent range for any A, Z range can be obtained, with a wide range of isotopes for which the efficiencies are around or higher than 5%. ECRIS breeders cover masses above 20 while EBIS systems cover the whole chart of isotopes.

- The charge breeding times are well below one second (whatever choice of charge breeder), which is shorter than or similar to the typical diffusion-effusion times from ISOL targets. Very short breeding times for short lived isotopes, down to the ms region with an EBIS breeder, are a priori possible.

- Intense radioactive beams - up to 1E13 ions/s - can be charge bred without loss of efficiency by ECRIS charge breeders.

- Exotic beams of medium to low intensity – as low as 1E2-1E3/s – can be charge bred by the EBIS keeping very good beam purity.

- CW operation of the superconducting LINAC will be possible using the natural mode of operation of the ECRIS charge breeder, and two EBIS charge breeders in "push-pull" mode.

In the future, the performances of ECRIS and EBIS may overlap more and more. For instance, one should expect purified beams from ECR charge breeders and true CW operation from EBIS charge breeders. It is not yet clear where the exact frontier between the two techniques and their domain of application will be. The numbers given above are only indicative, and both techniques are expected to exhibit improved performances with time as experience is gained and numerous developing goals are achieved.

e. Beam manipulation and purification

Progress in this very challenging subject is essential for providing high quality beams to the users. Major progress is expected in Coolers and mass separators and Gas cells and traps within the HIE-ISOLDE design study phase.

The Isolde ion sources are heavily optimised for efficiency, the principal concern when studying exotic isotopes. It would be difficult to make any substantial reduction of the ion-source's emittance without having some significant effect on its efficiency. The solution is to employ a separate beam-cooler, downstream of the ion-source and upstream of the separator's dispersive sections. Isolde already possesses a beam-cooler based on a continuous-injection RFQ ion-trap. A low-pressure buffer gas inside the RFQ cools the beam and achieves an extracted beam emittance of $\leq 3\pi$ mm.mrad. However the existing cooler is situated downstream of the HRS, and therefore has no effect on the HRS performance. We propose to design a new cooler, based on the existing design, but adapted for operation as part of the HRS.

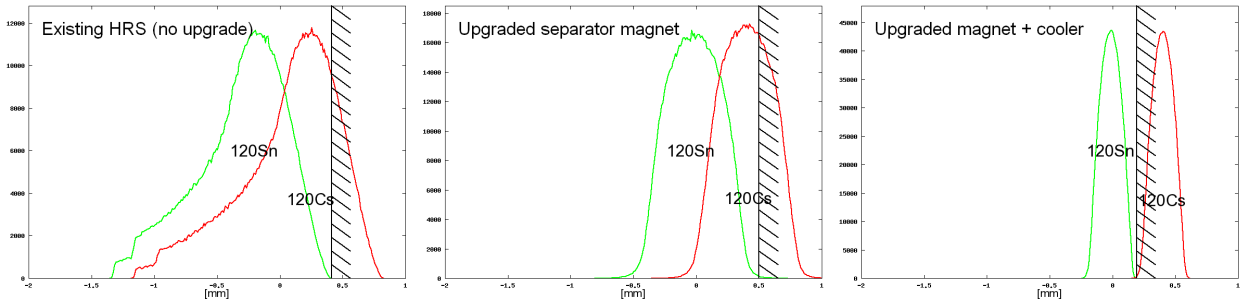


Figure 6: Example calculated beam profiles of the HRS

A calculation of the beam-profiles for A=120 beams is shown in figure 6. This example represents a typical case in which a upgraded HRS could deliver improved ^{120}Ag beam quality by drastically reducing the transmission of the unwanted background beam (in this case ^{120}Cs). The black vertical line represents a movable slit, positioned to optimise the statistical significance of the signal beam delivered to the experiment. The leftmost graph shows the situation for the un-upgraded HRS: in this case there is little the separator can do to suppress the unwanted isotope. The centre graph shows the effect of upgrading the separator magnet. The rightmost graph shows the effect of combining an upgraded magnet with a beam cooler: here the resolving power of the HRS is dramatically improved.

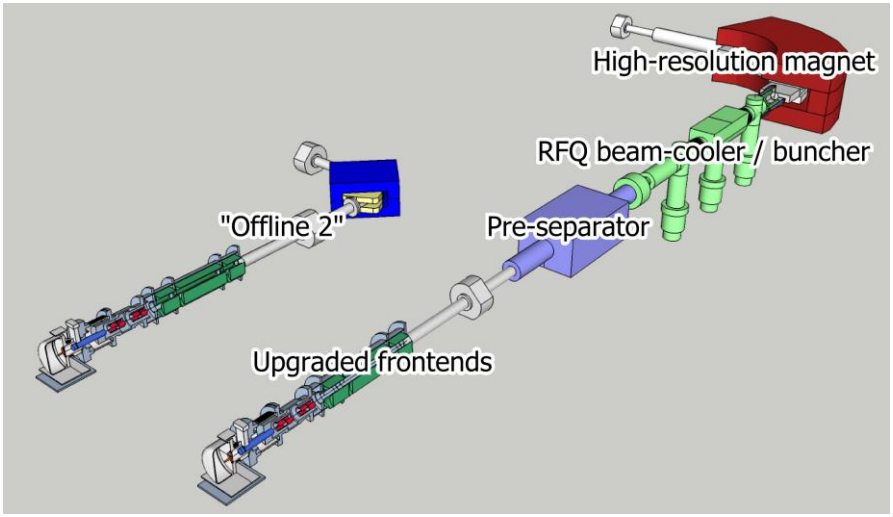


Figure 7: Layout of a Mark-II HRS offline test stand

To study the design and performance of a mark-II HRS we intend to construct an offline test stand, as illustrated in figure 6. Initially it will be a simple conventional low-resolution separator, to which prototypes of the cooler and corrected-magnet will be added as they become available. As shown in the diagram the cooler will also require a new pre-separator stage which must be designed and tested. The test stand will permit us to optimise the performance of each component and study its real-world performance with beam. It will also be used to verify the compatibility of each component and make adaptations where necessary. For example, the extraction stage of the cooler must produce a beam suitable for injection into the magnetic sections. There is limited space for dedicated matching sections, so it is desirable to design the cooler extraction optics to avoid the need for extra quadrupole lenses. The goal is to build a simplified but working version of the upgraded HRS, before any installation work begins on the Isolde facility.

5. Collaborations / Resources

The concentration of ISOL facilities in Europe is unique in the world, and coordination of their development work will strengthen Europe's technological excellence. All major European ISOL installations that are research infrastructures (ALTO, EXCYT (LNL-LNS), SPIRAL (GANIL), ISOLDE and IGISOL (JYFL)) are helping to lay the groundwork for the future "ultimate" ISOL facility EURISOL which aims to integrate all European ISOL activities at the horizon of the 2020 decade. EURISOL will encompass the progress being achieved today in many areas at the existing facilities, R&D which will be performed at the intermediate generation facilities (HIE-ISOLDE, SPES and SPIRAL2).

EURISOL related activities are overseen by the EURISOL Project Office (PO) which has four members: Yorick Blumenfeld (CERN, chair), Albeto Facco (INFN- Legnaro), Marek Lewitowicz (GANIL) and Piet Van Duppen (UC Leuven). The PO meets at least 4 times a year by telephone. Currently a EURISOL collaboration is being put in place which will have a steering committee representing the stakeholders.

Future EURISOL users are members of the EURISOL USER Group, which has over 1000 members. This group elects an executive committee of 9 members, renewed by half every two years. The current chair is Angela Bonaccorso (INFN-Pisa).

Within the ENSAR (European Nuclear Structure and Application Research, FP7) initiative funded by the European Commission, EURISOL-NET is led by Yorick Blumenfeld (IPN-Orsay) and comprises two tasks:

- Coordinating EURISOL R&D at current European ISOL facilities led by Yacine Kadi (CERN)
- Updating the EURISOL Physics case led by Angela Bonaccorso (INFN-Pisa)

EURISOL-NET will organize the next EURISOL town meeting in Orsay (October 2014) in collaboration with the PO and the User Group. The total budget of EURISOL-NET is 283 KEuros over 4 years.

Within ENSAR two Joint Research Activities (JRA), ACTILAB (led by Thierry Stora – CERN) and PREMASS conduct R&D for actinide targets and beam preparation respectively, useful for EURISOL.

The objective of the work package TIPHAC of the TIARA preparatory phase (FP7) is to coordinate the design of test infrastructures for two major technical issues before launching the construction of EURISOL: the development of high power targets (led by Yacine Kadi – CERN) and low beta superconducting accelerating structures (led by Sebastien Bousson – IPN Orsay). The total budget of TIPHAC is 690 KEuros over 3 years.

The EMILIE project, led by Pierre Delahaye (GANIL) and promoted by the FP6 network NuPNET, is a consortium of 9 European laboratories including CERN as associate partner which

conducts R&D on improving performances of Charge Breeders which is a critical issue for post accelerating EURISOL beams. The total budget of EMILIE is 513 KEuros over 3 years.

Thierry Stora (CERN) is leading an effort to build and irradiate a Pb-Bi loop at ISOLDE, which can be considered a prototype for the EURISOL converter target. This collaboration includes CERN/ISOLDE, CEA-Saclay (SPhN), ESS, MYRRHA, PSI, and Saha Institute Kolkata, India. CERN has committed 300 KCHF over 3 years and 2 FTE manpower to this R&D effort which exploits the synergies of the major European high power accelerator projects (ESS, EURISOL and MYRRHA).

6. Conclusions

An ultimate ISOL facility such as EURISOL holds broad scientific promise but represents a formidable technological challenge. Therefore a European roadmap has been established which includes building of 3 so-called “intermediate generation” ISOL facilities: HIE-ISOLDE which is a major upgrade of the current ISOLDE facility at CERN, SPES in Legnaro, Italy and the most ambitious which is SPIRAL2 at GANIL, Caen, France. These facilities, which are precursors to EURISOL will provide a ground for development and testing of many of the technological solutions outlined in the reports from the EURISOL Design Study.

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ANNEXES

1. The SPIRAL2 RFQ

EURISOL NET Meeting, 2013/05/28-29, Jyväskylä

Alain C. France, Vincent Hennion, Philippe Galdemard, Olivier Piquet, CEA/IRFU in Saclay

Introduction

The SPIRAL2 RFQ is designed to accelerate at 88 MHz two kinds of charge-over-mass (Q/A) particles, namely a 5 mA deuteron beam (Q/A = 1/2) at 3 MeV energy, or a 1 mA ion beam with Q/A = 1/3 up to 0.75 MeV/A energy. This CW machine has to show stable operation, provides high availability, and has minimum losses to minimize activation. We report here design studies (conducted by CEA/IRFU in Saclay) and construction status.

SPIRAL2 RFQ design

RFQ design generally follows a five-step process.

Step1. Beam dynamics study yields main RFQ parameters, as frequency (88.0525 MHz), length (~5 meters) and geometry of the axial region of the RFQ: electrodes tip-to-tip distance, electrode tip radius and electrode modulation. Recall that electric component of RF field is used both for particle bunches focusing (in transverse and longitudinal directions) and acceleration. Geometrical parameters and inter-electrode voltage are continuously varied along the RFQ as the result of an optimization process. SPIRAL2 voltage function (Figure 1) reflects this optimization process, with essentially bunching in the first two meters, and increased acceleration in the next three meters.

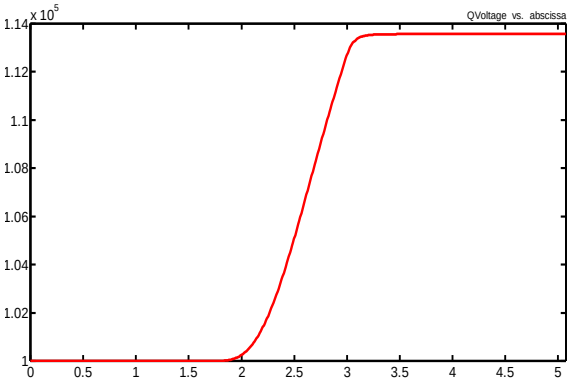


Figure 1. Inter-vane voltage (V) vs. abscissa (m).

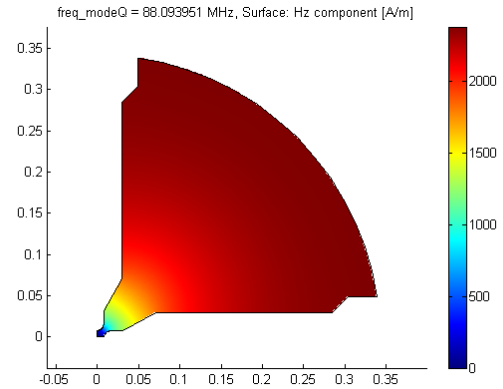
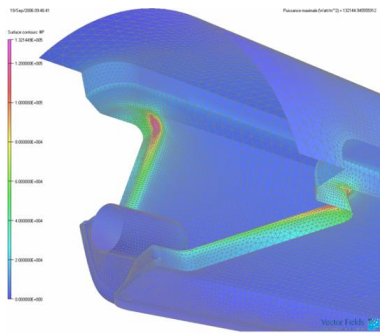


Figure 2. Magnetic field in RFQ cross-section.



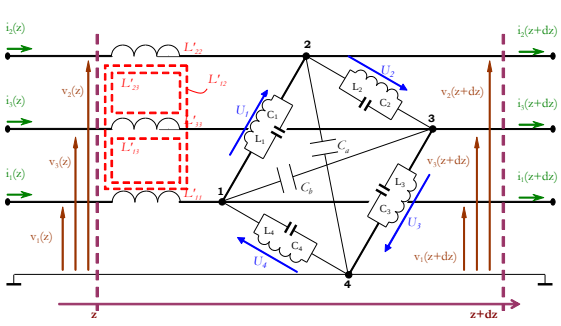
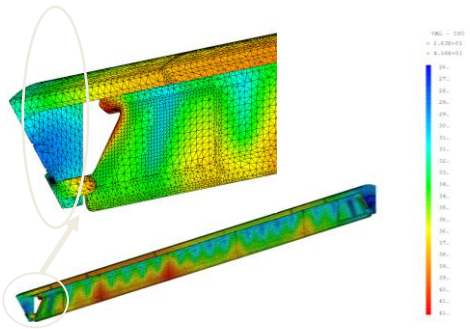


Figure 3. 3D simulation of RFQ end-circuit.

Figure 4. 4-wire transmission line model.



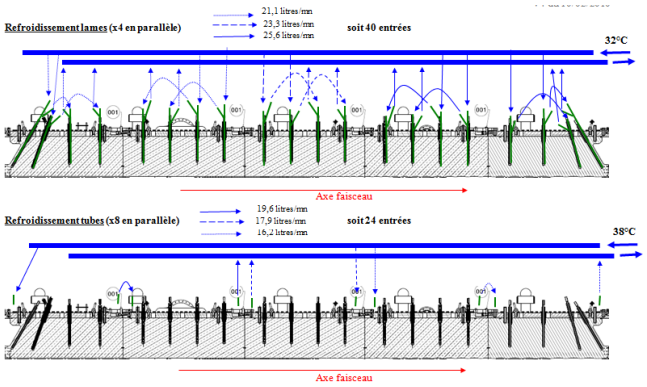


Figure 5. RF induced heating.

Figure 6. Water cooling channels.

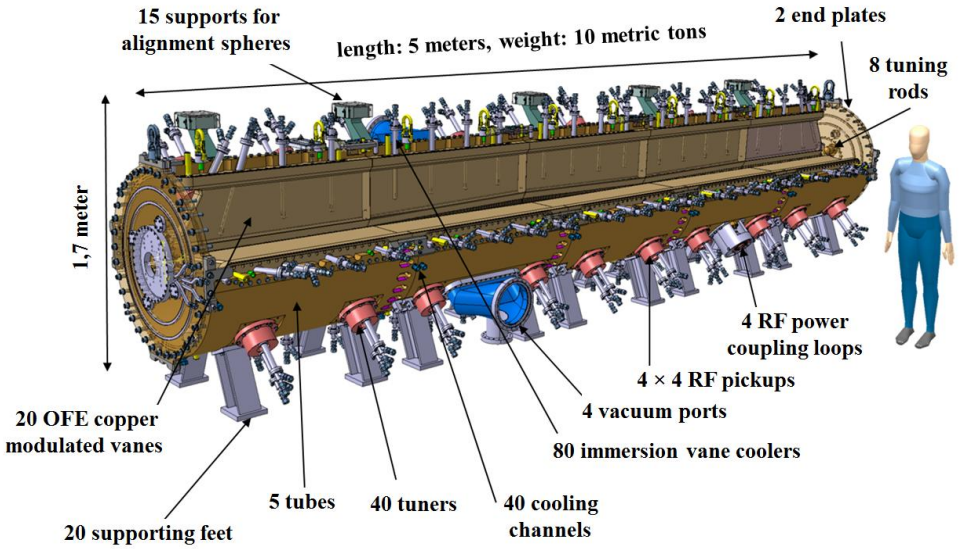


Figure 7. The SPIRAL2 RFQ (tube is opened along one quadrant).

Step 2. 2D and 3D RF calculations are then used to define detailed geometry of the cavity, cross-section (Figure 2), end-circuits (Figure 3), tuning slugs, RF power couplers and power dissipation. Electrical parameters of RFQ 4-wire transmission line model (TLM - model of an infinitesimal length of RFQ is shown in Figure 4) are also derived from 2D and 3D simulations, in such a way the model perfectly mimics the physical RFQ. TLM may be then safely used in error analysis and slug tuner dimensioning. TLM is also the theoretical background of RFQ tuning algorithm, as it establishes a bridge between 3D field maps of desired cavity and measurable quantities which are field profiles along bead-pull lines and frequency spectra.

Step 3. The so-called "RF stability", i.e. the sensitivity of RFQ to construction imperfections and to thermal deformations induced by power operation, is then evaluated. This allows assigning construction tolerances to the design, including machining errors of individual RFQ components and assembly errors. Most stringent tolerances apply to axial region, where typically some given relative error on electrode position may yield a ten times larger relative error on inter-electrode voltage. Effects of construction imperfections are compensated by a careful adjustment of 40 tuning slugs; tuning range extends up to 140 mm inside cavity with the specified machining/assembly tolerance envelope.

Step 4. 3D thermo-mechanical simulations are used to estimate deformations induced by RF heating (Figure 5), and the cooling system is designed to maintain geometry within bounds compatible with the RFQ specifications, mainly ±1% voltage error and a few tens of kHz detuning. Two independent cooling systems

(Figure 6), respectively for the tube and the vanes, are temperature-controlled with 0.1°C accuracy to this purpose. Note that water circulation is alternated from one-meter long module to the next, in such a way deformation amplitude exhibits a sawtooth profile hence minimizes voltage error.

Step 5. The machining and assembly processes satisfying tolerance requirements are defined, and detailed drawings are edited. A 3D view of the RFQ is displayed in Figure 7.

RFQ construction and integration in GANIL

RFQ machining and assembly is realized in Research Instruments GmbH. Each 1-meter long RFQ module is made of one 800 mm diameter copper tube and four copper electrodes. After CMM control of individual parts, electrodes are successively fitted with vacuum and RF seals, inserted inside the tube using dedicated apparatus, and bolted into position (Figure 8). Final acceptance is subjected to successful CMM control and vacuum test.

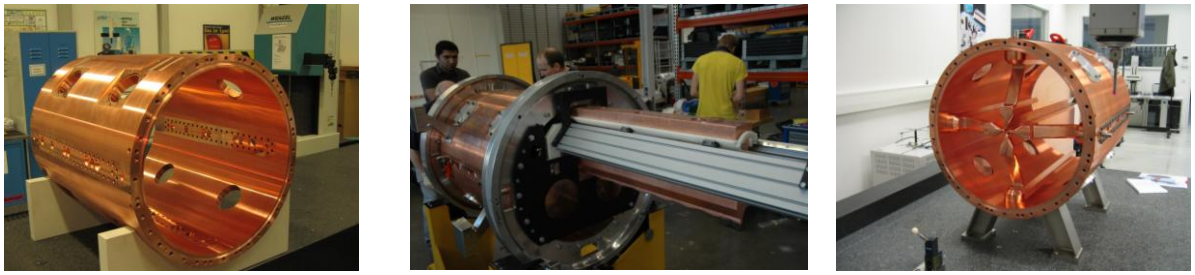


Figure 8. From left to right: a copper tube, inserting an electrode in a tube, a completed module.

Integration in GANIL requires study, specification and realization of numerous ancillaries, including

– handling tools useful for assembly, tests and transport of RFQ modules,

– RF tuning bench using bead-pull perturbation method for field probing,

– cooling and temperature control systems,

– low-level RF (LLRF) systems, designed to regulate RFQ voltage via control of RF amplifiers output power and temperatures of the two cooling circuits (tubes and vanes),

– four 60 kW RF power amplifiers (under GANIL procurement),

– vacuum systems (under GANIL procurement).

Integration starts with assembly of the five modules on their support and mechanical alignment (using standard optical methods). Installation of cooling systems is started; a minimum functionality is required at this step, since RFQ just needs to be temperature-stabilized for RF tuning. RF tuning is performed next: specified voltage profile and cavity resonance frequency are obtained via position adjustment of the 40 tuning slugs. RF and vacuum systems are then installed, and RFQ conditioning may start. RFQ voltage is progressively ramped up to nominal value; parameters of the two temperature control loops and LLRF systems are adjusted. RFQ is commissioned after output beam measurements.

Status as of May 2013

Expected timeline for RFQ modules delivery is given in

Table 1. T5, T4 and T3 modules are following the acceptance process. T2 and T1 modules are in final machining state. All five modules are expected to be ready for assembly in GANIL at end of Sept. 2013. Note that T4 and T3 will be first shipped to Saclay for preliminary tests:

– the assembly procedure will be tested with T4 and T3;

– the RF tuning bench will be tested with T4, and eventually with T4-T3 assembly in case enough time is left; RF parameters of these two modules will be estimated by the way;

– the low-level RF system will be tested with T4.

Assuming all modules to be available at GANIL at end of Sept. 2013, RFQ commissioning should start at beginning of July 2014, after 2-month assembly, 4-month tuning, and 2-month conditioning (Table 2).

Table 1. Expected timeline of RFQ modules delivery.

Module	Operations	Expected timeline
T5	Leak test with new vacuum seals	May 27~31, 2013
	CMM	
	Delivery in GANIL	July 31, 2013
T4	CMM in progress, expected to be within tolerances	
	Delivery in Saclay	June 15, 2013
T3	Assembly and leak test	May 27 ~ 31, 2013
	CMM	
	Delivery in Saclay	July 31, 2013
T2 & T1	Vane final machining	July 15, 2013
	Assembly/vacuum test/CMM	
	Delivery in GANIL	September 30, 2013

Table 2. Expected timeline of operations in GANIL.

Operations	Expected timeline
arrival in tunnel	October 01, 2013
modules assembly and alignment	October 01 to November 30, 2013
RF tuning	December 01, 2013 to March 31, 2014
RF conditioning	June, 2014
beam tests	starting on July 01, 2014

2. *Multi-reflection time-of-flight mass analyzer for analysis, optimisation, mass separation and precision mass measurements of radioactive ion beams*

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Multi-reflection time-of-flight (MR-ToF) mass analyzers are among the most recent developments in mass spectrometry and manipulation of low energy (keV) ion beams. They offer highest mass resolving power on the order of $R=m/\Delta m$=100,000 on short time scales of a few tens of milliseconds. Therefore, they are ideally suited as, e.g., mass analyzers of short-lived ion beams, mass purifiers in combination with a fast ion selector and high-precision mass spectrometers. Although conceptually developed and tested already in the early 1990s [1], the application to radioactive ion beams was achieved not until recently. Several groups have developed MR-ToF devices for nuclear physics investigations; the SLOW-RI team at RIKEN [2-4], the FRS-ioncatcher group at GSI [5-7] and the ISOLTRAP collaboration at ISOLDE/CERN [8,9]. Furthermore, this type of device is under development at many other radioactive ion-beam facilities [10,11]. In the following, the design and achievements in performance and application versatility of the MR-ToF mass analyzer installed at ISOLTRAP/CERN will be presented.

Figure 1 shows a sketch of this MR-ToF mass analyzer, visualizing its principle of operation and the two general application modes. An ion bunch of short time distribution, typically some tens of nanoseconds – $\Delta t_{FWHM}\approx$60ns in the case of ISOLTRAP where it comes from a radio-frequency-quadrupole ion-beam buncher and cooler (in short RFQCB) with a mean kinetic energy of 3.1keV – is injected into the MR-ToF mass analyzer [12]. The analyzer consists of two electrostatic ion mirrors between which the bunch is electrostatically confined in radial and axial direction. The injection and ejection is achieved via an in-trap lift electrode which facilitates the operation and the performance of the analyzer [13]. Due to the folded flight path of the ion bunch of up to several kilometres, which corresponds to flight times t of several tens of milliseconds, high mass resolving powers $R=m/\Delta m=t/(2*\Delta t)$ on the order of 100,000 can be reached, sufficient to resolve most isobaric ions. Several of a broad range of possible applications are described in the following.

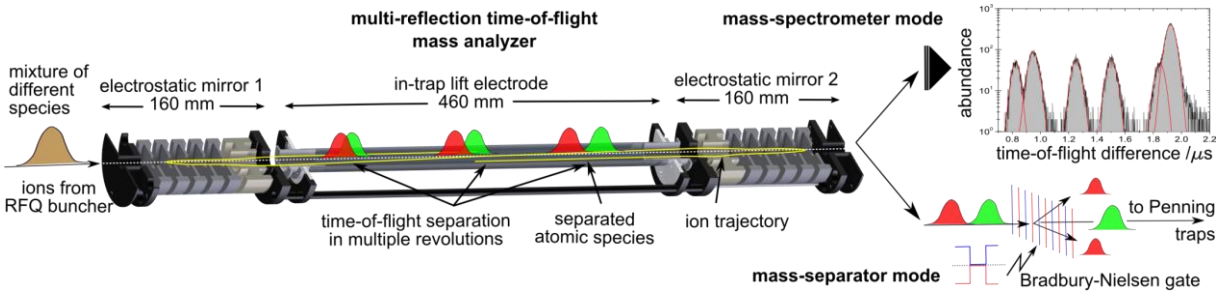

Fig. 1: Sketch of the operation principle of the ISOLTRAP MR-ToF MS as a mass analyzer and precision spectrometer in combination with an ion detector (top right) and as a mass separator in combination with a Bradbury-Nielsen beam gate (bottom right) [9].

High-resolution ion beam composition identification, analysis and monitoring for various ion-beam production and ionisation developments and characterisation

Figures 2 and 3 show examples of applications where the MR-ToF mass analyzer was used to characterise the influence of different parameters affecting the radioactive ion-beam production. Due its fast (milliseconds) cycle, non-scanning mode and high mass resolving power, the direct evolution of the yield of the different species can be recorded. In Fig. 2, the target temperature was increased which resulted in a yield increase for short-lived copper isotopes. Due to the complexity of the delivered beam, containing short-lived nuclides, stable nuclides and molecular sidebands on this particular mass number, other beam-analysis techniques such as Faraday cups or decay stations fail to detect the full composition qualitatively and quantitatively [9]. In Fig. 3, resonant laser ionisation (RILIS) for short-lived Au isotopes was used to produce a gold beam, which was contaminated by the corresponding Tl isotopes. The Au component can be unambiguously identified by switching off the laser. The evolution of the Au yield was monitored with the MR-ToF mass analyzer by gating only on the Au isotopes and scanning the first RILIS excitation step. This resulted on the one hand in the optimized excitation wavelength for this particular atomic transition and on the other hand revealed the isotope shift and the hyperfine structure of the isotope. In comparison to Faraday cups, this method is orders of magnitude more sensitive and quasi background free [9,14]. In contrast to yield measurements with a decay station, it is independent of the branching ration and not limited by an upper half-life. A third example, shown in Fig. 3, is the sampling of the release curve of a particular species from the target and ion source system. Due to the MR-ToF MS' short experimental cycle time of only a few milliseconds, even these fast processes can be monitored and optimized. By varying the delay after each proton pulse before ion injection into the RFQCB, and subsequent yield analysis, the effusion and diffusion times of the target can be evaluated [15].

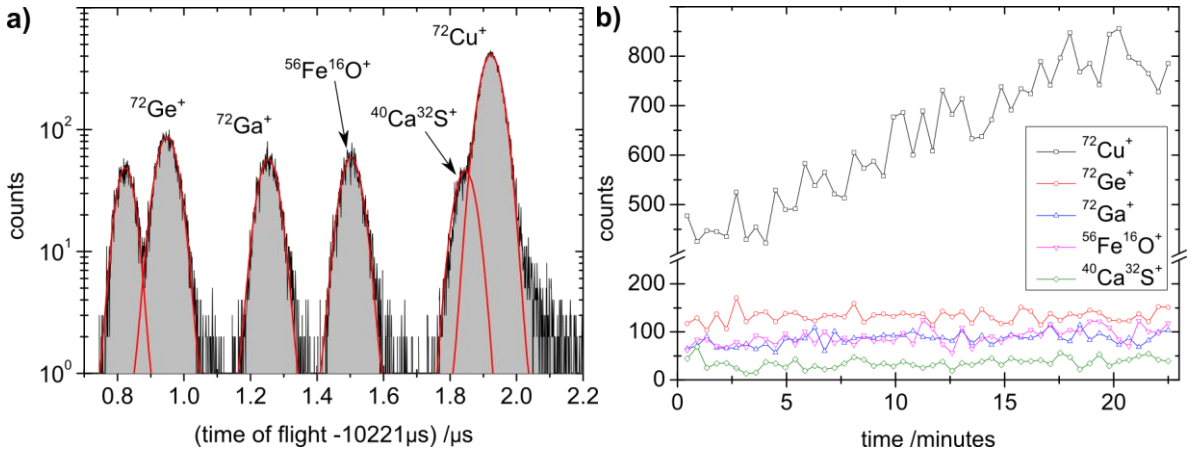

Fig. 2: a) Integral time-of-flight spectrum for $A=72$ as delivered from ISOLDE during heating of the target material; b) Number of counts of the isotopes from the spectrum on the left as a function of the time and increase of the target heating by approximately 70°C [9].

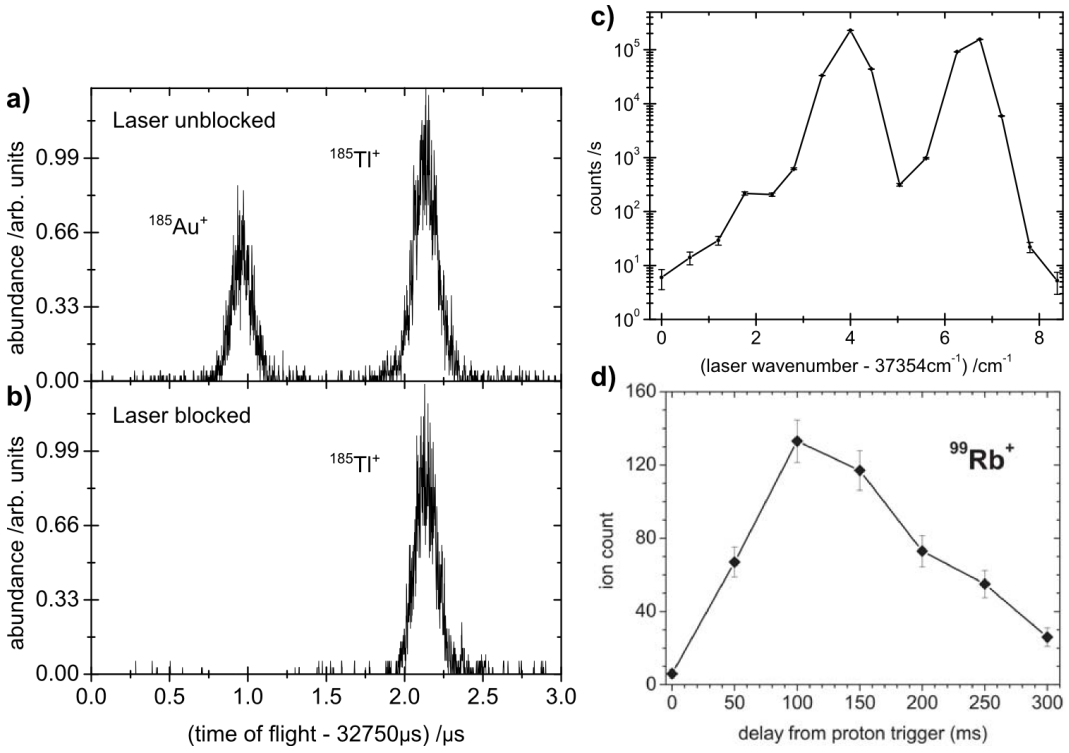

Fig. 3: left: $A=185$ time-of-flight spectra after 1500 revolutions in the MR-ToF MS. a) The RILIS lasers were tuned to ionize $^{185}\mathrm{Au}^{+}$ atoms. The clear separation from $^{185}\mathrm{Tl}^{+}$ isotopes enables a background-free gold-yield analysis. b) Furthermore, blocking the laser beam unambiguously identifies the gold isotope. [9] c) Signal intensity of $^{185}\mathrm{Au}^{+}$ (scaled to RFQ accumulation time of $1\mathrm{s}$), representing the ionization efficiency, as a function of the wavenumber of the RILIS first excitation step ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$; the two resolved resonances of the hyperfine-structure correspond to the transitions $F=3\rightarrow 2,3$ (left peak) and $F=2\rightarrow 2,3$ (right peak)) . The laser line width was $0.33\mathrm{cm}^{-1}$. [9] d) Scan of the target and ion source release curve by varying the delay between the proton irradiation of the target and the start of the RFQCB ion accumulation. [15]

MR-ToF mass purification

In combination with fast ion deflector, e.g. a Bradbury-Nielsen gate [6,16], the MR-ToF separated species can be selectively addressed for further transfer to a subsequent experimental step. So far at ISOLTRAP, a contamination suppression of four orders of magnitude has been reached [12]. The purified bunches can, e.g., be used to supply Penning traps for high-precision mass measurements [17] or any other investigation. For example, the MR-ToF MS will offer a fast alternative for trap-assisted decay-spectroscopy experiments at ISOLTRAP, where currently the purification is performed by Penning-trap mass separation [18].

MR-ToF mass measurements

The MR-ToF device can also be used as a mass spectrometer of its own right which is very fast and sensitive, i.e. can be applied to nuclides of short half-lives (down to a few milliseconds) which are available with only very low yields – even a few counts of the ions of interest are sufficient. As most often, signals of other isobaric species are found in the time-of-flight spectra, these provide ideal (because close-lying) reference mass values for the calibration. Thus, mass measurements with low uncertainty, i.e., $\delta m/m \approx 10^{-7}$, are possible. This opens the possibility to explore the so far unknown masses of nuclides on the outskirts of the chart of nuclides. Particular applications are mass measurement in regions approaching the r-process path, rp-process path or nuclides relevant for neutron-star crusts. A recent first application was the mass measurement of the neutron-rich calcium isotopes ^{53,54}Ca [19].

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3. TWIN – Thermal-hydraulic experiment of the heat transfer coefficient on a concave beam WINdow

EURISOL-NET meeting Jyväskylä May 28th -29th 2013

K. Samec & Y. Kadi, CERN

Objectives:

A key component of a 4 MW / 1 GeV / 4 mA high-power neutron spallation target; an innovative concave beam window design has been tested in conditions hydraulically representative of full power in the EURISOL project, supported by the FP7 program. The new high-power target design is characterised by a thin-gauge concave beam window optimised to withstand very high pressure and thermal stresses from a 4 MW proton beam converter able to emit a very dense neutron flux in the order of 10^{15} n/cm^2/s. The hydraulic test confirmed that despite a thickness as low as 0.8 mm in places, the beam window could sustain a high-velocity dense liquid-metal flow able to absorb the 4 MW beam over a 15cm diameter compact spallation target. Hence, there is sufficient credible data to predict that such a high-power target would work under beam. The lifetime of the beam window depends critically on the local heat transfer coefficient which has yet to be measured. The current proposal aims to use a robust accurate strain gauge technology to measure heat transfer coefficients in a thermal-hydraulic test of the EURISOL beam window and thereby validate the design of the innovative beam window for use under beam. Such a graded approach minimises development risk and would benefit future high-power targets.

Prior experience:

The design shown in figure 1 below was built at full scale and tested hydraulically at a flow rate of 150 kg/s, sufficient to evacuate the required beam power. The liquid metal was monitored and pressure fluctuations were found to correspond to calculations performed using Computational fluid dynamics as shown in Figure 2. However the test did not include beam irradiation at 4 MW as no such facility yet exists although one is being envisage at ESS in Lund. The team therefore propose to test the thermal characteristics of the beam window using another method, to demonstrate the capability of the design to handle 4 MW.

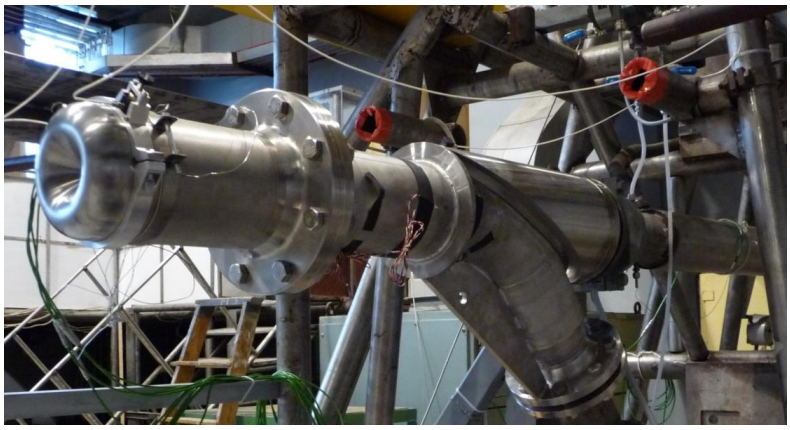

Figure 1: EURISOL Liquid metal neutron converter on test stand an IPUL

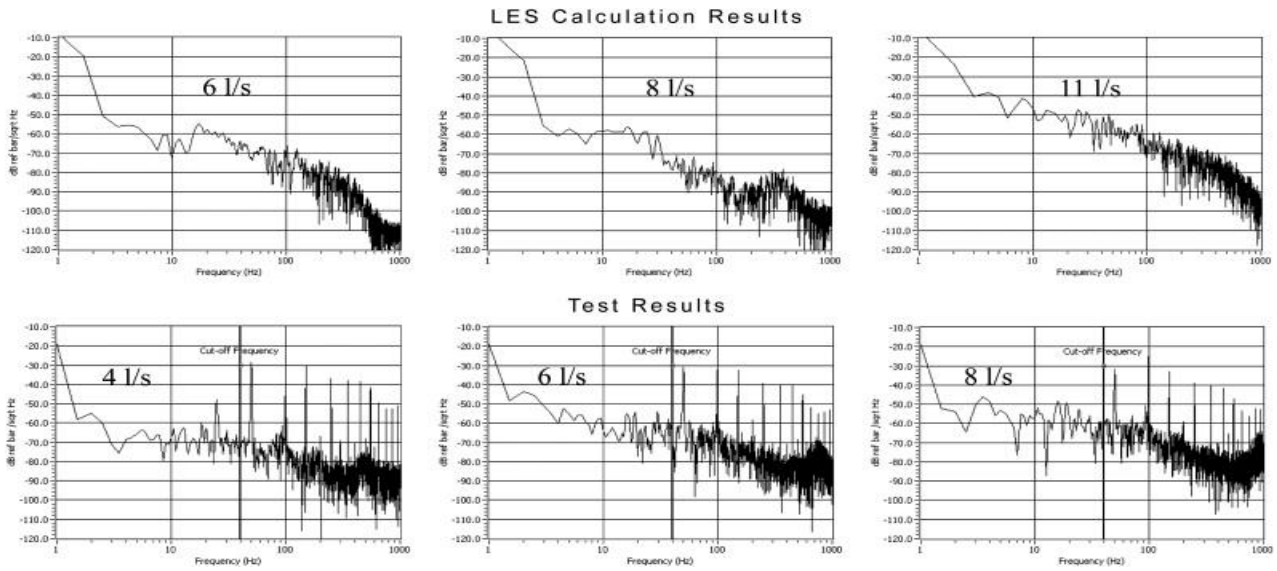

Figure 2: Comparison of pressure fluctuation spectral densities calculated (top) and measured (bottom) in the hydraulic test

Proposed experiment:

In a first experiment the strain gauges would be tested on LBE-wetted bimetallic samples placed in a pressurised heated chamber, filled with liquid metal. The goal is to validate the strain gauge concept for measuring the temperature of a surface wetted by liquid metal and verifying its durability and robustness. The thermal strain will be measured and compared against a thermocouple reading to validate its accuracy at temperatures up to at least 350°C and beyond, if possible.

Once the measurement technique has been validated, it shall be used to measure the heat transfer coefficient on the new concave beam window used in the original EURISOL project under the same full-power hydraulic conditions as in the original test to validate experimentally the ability of the window to sustain the impact of a 4 MW beam. The first EURISOL test was purely hydraulic and has already validated the hydraulic design. Validating the thermal design of the window is the next logical step.

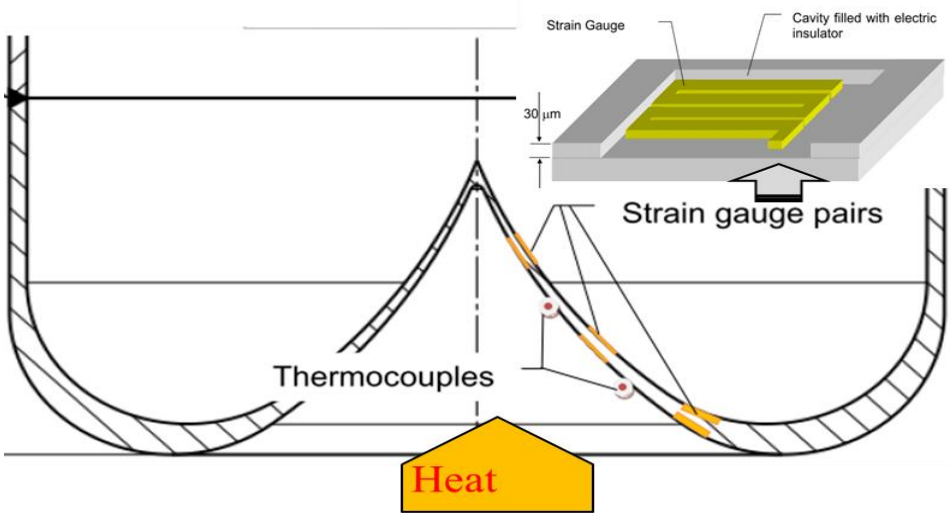

Figure 3: Schematic of beam setup on beam window

Relevance:

The proposal relates specifically to the development of high-power neutron sources, liquid metal technology, ADS energy systems. The instrumentation used is relevance to heat flux measurement in high-temperature, corrosive and electrically conductive liquids.

Infrastructure:

The Eurisol target from the EURISOL-DS (FP7) test in 2008 may be re-used along with the facilities at IPUL, reducing costs and supporting Infrastructure in a new EU-8 country. The strain-gauge technology will be developed with an established Swiss private company making use of existing technology and infrastructure used in the past for the MEGAPIE project (FP6).

Involved Partners:

CERN (Geneva, Switzerland) and IPUL (Latvia) are the participating institutes

Outcomes:

The expected outcome of the project is a validation of the new EURISOL beam window for high-power (4MW) spallation sources. An additional benefit would rest in the development of a robust heat flux measurement techniques suitable for liquid-metal that may be applied also to other high-temperature opaque fluids.

Budget:

Requested material+ Travel Costs:	295 k€
Requested manpower Costs:	285 k€
Total Costs:	580 k€

Deliverables:

- Instrumentation Report — Month 12
- Test Plan — Month 24
- Project Report — Month 48

4. *Recent ISOLDE RILIS developments and outlook for laser ionization at next generation ISOL facilities.*

V.N. Fedosseev, CERN

The Resonance Ionization Laser Ion Source (RILIS) of the ISOLDE on-line isotope separator at CERN is based on the method of laser step-wise resonance ionization of atoms in a hot metal cavity [1, 2]. Two or more laser beams, wavelength-tuned to atomic transitions, resonantly interact with radioactive atoms inside a tubular hot cavity connected to a target. The step-wise excitation and ionization of the atoms is provided by high repetition rate lasers. Each laser wavelength is precisely tuned so that the photon energies match the unique combination of successive electronic transition energies within an ionization scheme for a chosen element. The main advantages of resonance ionization laser ion sources are the chemical selectivity of ionization and a convenient means of manipulating the ion beam composition, simply by blocking or unblocking the laser beams. The RILIS laser installation is located in the ISOLDE experimental hall in a position which allows transporting the laser beams to the front-ends of both ISOLDE mass separators.

The RILIS installation at ISOLDE has been constructed in beginning of 90ths and is used for physics experiments since 1994. Originally it was based on dye lasers pumped by high repetition copper vapour lasers. A staged upgrade of the RILIS installation has been performed in the period of 2008-2012 [3]. It included replacement of copper vapour laser by solid-state Nd:YAG lasers, replacement of old home-made dye lasers by new commercial dye lasers and, finally, installation of complementary solid-state Titanium Sapphire (Ti:Sa) lasers. With the use of harmonics generation in non-linear BBO crystals, the tuning range of RILIS lasers covers the range of 210-945 nm without gaps. The both Ti:Sa and dye lasers are capable to generate pulses synchronously. This allows using them in different combinations optimized for each particular case. Thus, new modes of RILIS operation have been realized which profited for an increase of RILIS ionization efficiency and for faster switchover of RILIS setup between different ion beams.

The main directions of present and future RILIS development are: extension of the range of available ion beams, increasing the ionization efficiency and improvement of selectivity. Due to the RILIS upgrade a substantial progress has been achieved in first two directions. A tremendous improvement of ionization selectivity has been reached via implementation of the Laser Ion Source Trap (LIST) in collaboration with Mainz University. Although the ionization efficiency with LIST was found 20 times less than that of RILIS, the suppression of surface ions by up to 4 orders of magnitude was demonstrated [4]. This enabled in-source spectroscopy study of polonium isotopes which were heavily contaminated by francium isobars in conditions of the standard RILIS approach.

The work on improvement of LIST performance is continuing. In particular, the observed selectivity reduction for certain isotopes caused by in-trap ionization via nuclear decay is addressed by using thinner rods for the LIST rf-quadrupole structure. The study of the time structure of LIST ion beams is on-going at Mainz University. Gating of the ion beam synchronously with the laser pulses would further increase the selectivity of ionization.

The experiments with LIST coupled to UCx-target made evident the effect of high ion load on the efficiency of ion extraction from the hot cavity. This is to be taken into account for considering an optional use of the LIST in the ion-guide mode when the laser ions created inside the hot cavity (as in the standard RILIS scheme) are extracted. However, the LIST mode of operation should provide an efficient extraction of ions created inside the LIST RFQ structure.

In the standard RILIS cavity the extraction is provided by the penetrating external high voltage field of the mass separator and by a weak gradient of the internal cavity potential due to the dc heating. Although at typical regimes of ISOLDE targets these fields seem sufficient to compensate the charge load effect, it could become a limiting issue for applications of RILIS (and surface) type ion sources at the high current ISOL facilities. Therefore the development of ion sources with higher internal filed is important. This can be achieved by constructing cavities with higher electrical

resistance in the longitudinal direction. In the past, RILIS ion cavities made of thin niobium foils were successfully used in experiments at ISOLDE [5, 6]. Due to the higher resistance the electrical filed required to reach working temperature of the cavity was higher and the ion bunch of laser ions could be compressed. This provided conditions for effective use of gating the ion beam synchronously with the laser pulses (micro-gating) and attaining in this way an additional suppression of isobaric isotopes ionized continuously by the hot surface. Recently a high resistance ion source cavity made of crystalline graphite has been reported by V.I. Mishin [7]. The electrical resistivity of this material is changing by 3 orders of magnitude depending of the direction of applied voltage with respect to the crystalline structure. Thus, a substantial increase of the internal field gradient of the RILIS cavity can be achieved by using this material. It is expected that the efficiency of the RILIS with such high resistance cavity will stay high even at high ion current as the ions will be extracted much faster. In addition, the laser ion will form short (< 10 μs) ion bunches providing perfect conditions for selectivity improvement by the micro-gating technique. The R&D on high resistance graphite cavities for RILIS is started at CERN within the target and ion-source development programme.

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5. Status on Neutron Converter for SPIRAL2

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INFN Laboratori Nazionali di Legnaro, 35020 Legnaro, Italy

On behalf of the LEA_SPIRAL2 Collaboration

Abstract

The SPIRAL2 project is aimed to produce high intensity radioactive ion beams by adopting the most suitable method for each one of these desired radioactive beams. The radioactive ion beams (RIB's) will be produced by the "Isotope Separation On-Line" method (ISOL) via a converter (i.e. neutron converter) or by direct irradiation. Combination of both methods (i.e. via fission induced by fast neutrons in a Uranium Carbide target or by direct bombardment of the fissile material) it will allow covering broad areas of the nuclear chart. Moreover, it will allow carrying out promptly significant experiments and activities in both, fundamental and applied Nuclear Physics (medicine, biology, solid state, etc).

This document is describing the present status of the neutron converter, including the results of the R&D on the main critical components and maintenance tools.

Introduction

The neutron converter has to produce an intense flux of fast neutrons, mainly in the forward direction respect to the incoming deuteron beam, it must induce 10^{14} fissions per second all over the Uranium Carbide target, which is located upstream the converter. The primary beam consists of 40 MeV deuteron with maximum current up to 5 mA (200 kW).

The neutron converter is housed inside the production module and is physically separated from the Target Ion Source (TIS) by a stainless steel panel. During operation time, the production module is located in the production area, inside a shielded bunker. Production module removal must be done only by remote handling device. Disassembling, spare parts replacement, or conditioning of elements have to be conducted within the hot-cell to ensure that the radioactivity is confined. The current design integrating the neutron converter inside the production module is shown in Figure 1.

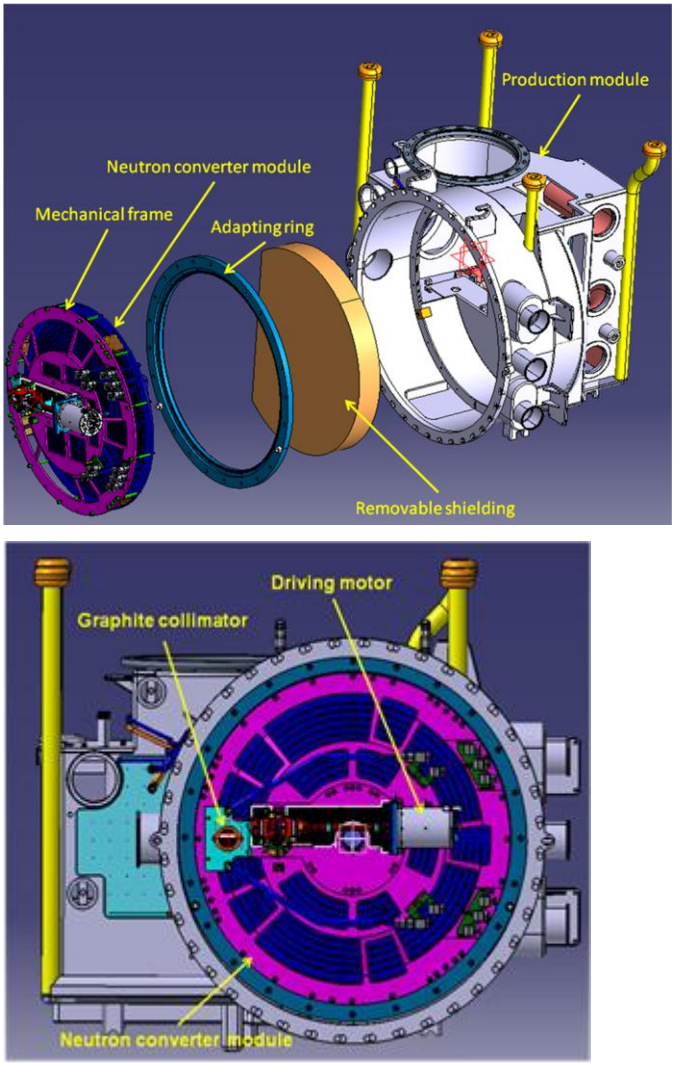

Figure 1. View of the neutron converter integrated inside the production module.

The neutron converter is conceived as a high speed (400 turns/min) rotating graphite wheel operating at peak surface temperature of 1850 °C.

Nuclear graphite made of natural carbon is a very suitable material as neutron converter. In fact, $^{nat}C(d,n)$ reaction is very prolific, especially in the forward direction where the neutron yield is comparable to that generated by other light material converters.

The thermal properties of graphite (melting point of 3632 °C) allow a compact geometry and the power dissipation from the converter does not demand a sophisticated cooling system. The heat is exchanged by thermal radiation with the water cooled panels. An electrical motor allows the graphite wheel to rotate.

At the beginning, while the performance is assessed, the facility will be operated at reduced power, up to 50 kW. The suitable neutron converter has been studied for this first period and has been designed based on the 70 kW prototype experience [1].

The design of the Neutron Converter Module (NCM) (mechanical frame, cooling system, delay window, remote handling,…) has been conceived to house both converters (50 and 200 kW), according to the user requirements. In practice, to switch the operative power is required to replace only the graphite wheel, not the module. The cooling panels, the delay window, the driving motor and the servitudes remain the same and may be re-used several times. Neutron converter has been conceived to operate as “nuclear device”.

The Neutron Converter Module (NCM)

The NCM consists of a mechanical frame which holds the neutron converter, the cooling panels, the graphite wheel, the delay window and the related servitudes (see Figure 2). The NCM is designed to satisfy the requirements of the “nuclearization”. Its installation, removal and maintenance must be done remotely by telemanipulator.

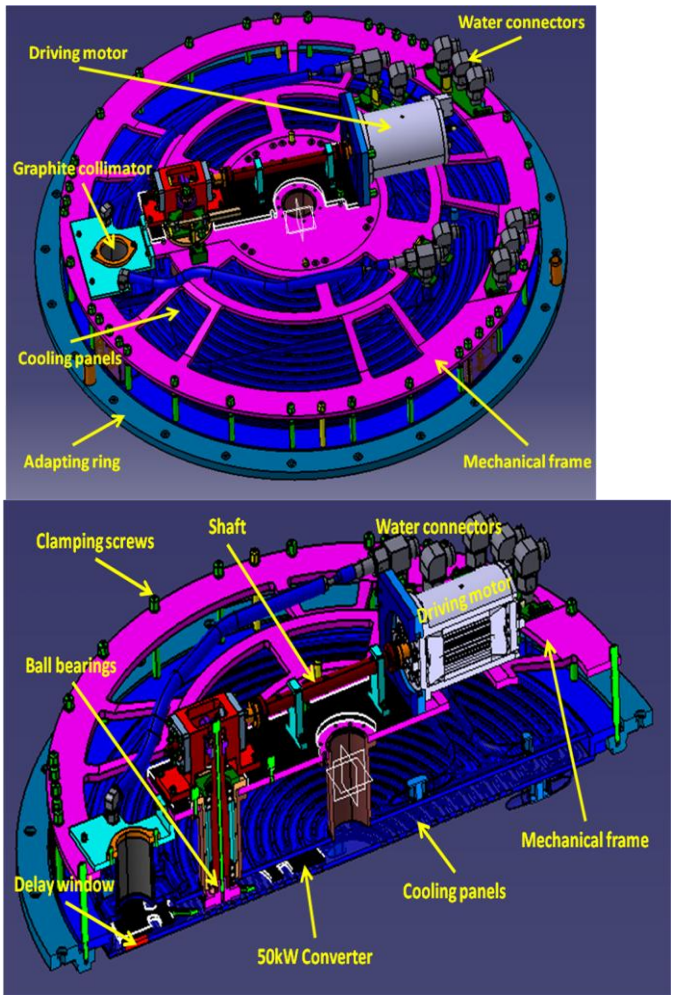

Figure 2. Left – A general view of the NCM. Right – a transversal cut of the NCM showing the internal components

At the working position the NCM is fixed on the mechanical adaptation ring with special clamp screws. In the very same way, the ring is fixed to the production module flange. The separation between the converter and the target-ion source is provide by two graphite gaskets, type CEFIGRAF from Garlock. A first gasket provides the tightness between the adaptation ring and the production module flange. A second gasket provides the tightness between the NCM and the ring.

The removable shielding allows the use of the NCM for different target configurations, by modifying the mechanical adaptation ring.

The active part of the converter is represented by a graphite rotating wheel (see Figure 3). The thickness of the graphite plates is 8 mm, well enough to stop the 40 MeV deuteron beam inside (stopping length 5.6 mm). The working temperature is up to 1850 °C and the disk is divided in sectors to compensate the graphite dilatation.

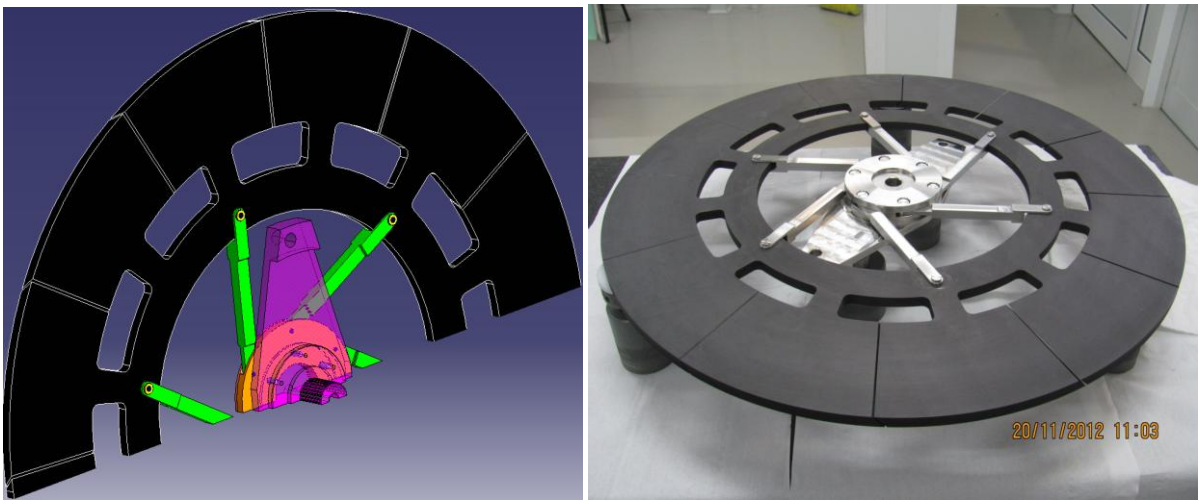

Figure 3. A conceptual sketch of the graphite wheel and a picture of the 50kW prototype.

The converter wheel is driven by an electrical asynchronous motor, housed under vacuum inside the production module and clamped to the mechanical frame of the NCM.

The driving electrical motor has been specially designed to operates under vacuum and radioactive environment [2]. Two couples of ball bearings specially designed to operate at high temperature, under vacuum and in absence of lubricant, assure the converter rotation [3].

The movement is transmitted to the converter through the transmission shaft (see Figure 4), which length varies on function of the converter's diameter.

Figure 4. Rotation system for the 50 kW converter. The system has been conceived to replace the block with bearings keeping the converter in place.

The correct rotation of the converter is controlled by monitoring three main parameters, independent from each other, i.e. voltage, motor absorbed current, and shafts rotation by inductive pickups located on the motor shaft and the converter shaft. In addition, a pair of thermocouples are monitoring the motor driver and the converter shaft temperatures.

A collimator is installed in front to the converter to protect the neutron converter from beam instabilities and define the right shape of the beam (see Figure 5).

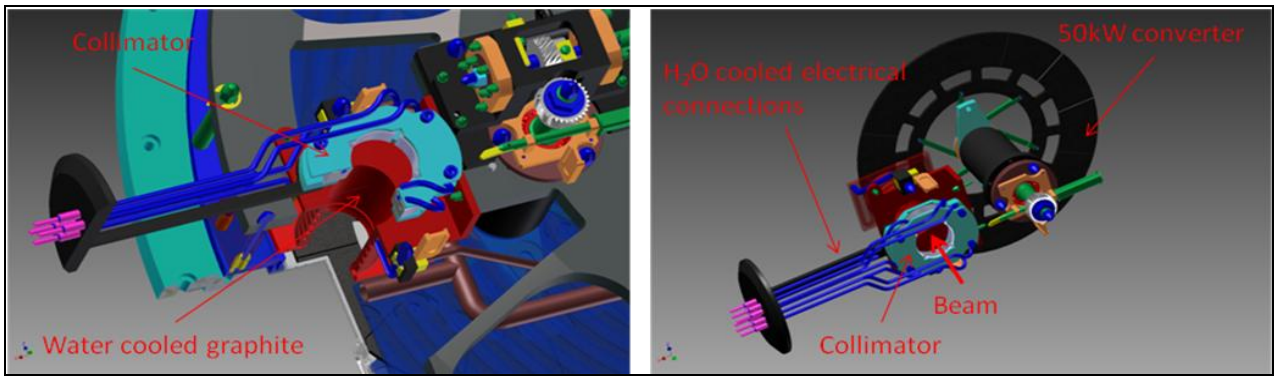

Figure 5. The production module with installed the beam collimator (left) and the set up for the 50 kW converter including the water cooled graphite tube (right).

The collimator consists of 4 sectors, electrically insulated from each other, mechanically mounted on the production module door. The collimator itself is made of molybdenum and the sectors are water cooled through tubular conductors connected externally by electrical feedthroughs. The measurement of the current on the sectors gives position to the beam and allows the continuous beam monitoring.

A water cooled graphite tube is placed right after the collimator, limiting transversally the beam path and collecting the back scattered electrons from the converter surface.

The "Delay Window" (DW) is a safety device designed for avoiding the direct interaction of the deuteron beam with the UCx target, in the case of failure of the NC and is located between UCx target and the rear part of the NCM (see Figure 6).

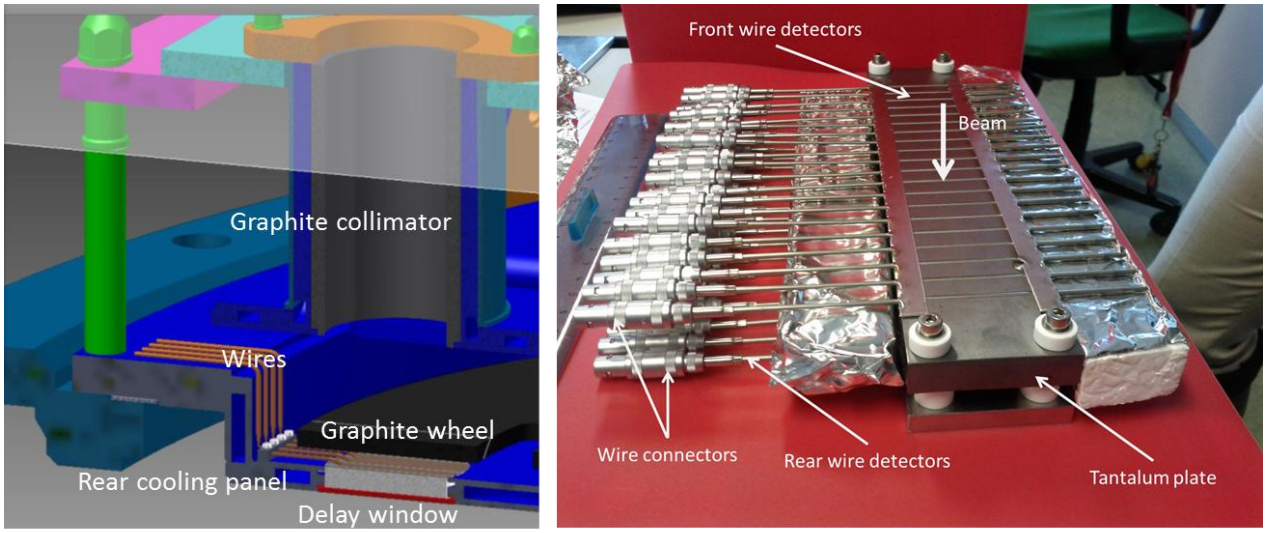

Figure 6. A sketch of the Delay Window (left) and the prototype tested with e-beam (right).

The DW operation consists in the absorption of the beam power (200kW) for a period of time longer than the time required for stopping the beam operation. The device consists of a tantalum disk, 12mm thick, equipped with a few electrical sensors (wires). The experimental results shown that the DW may withstand the beam power for a time of about 700ms, while the measured intervention time is only of 5ms [4].

Maintenance of the Neutron Converter Module

The maintenance of the NCM is planned to be done every three months of operation, according to the facility cycling. During the maintenance operations, some critical components of the NCM are integrally replaced while some others that show a longer lifetime are checked and hence reused in the next cycle of operation. Every single component of the NCM has been conceived to be handled by remote telemanipulators, inside the hot cell (Figure 7).

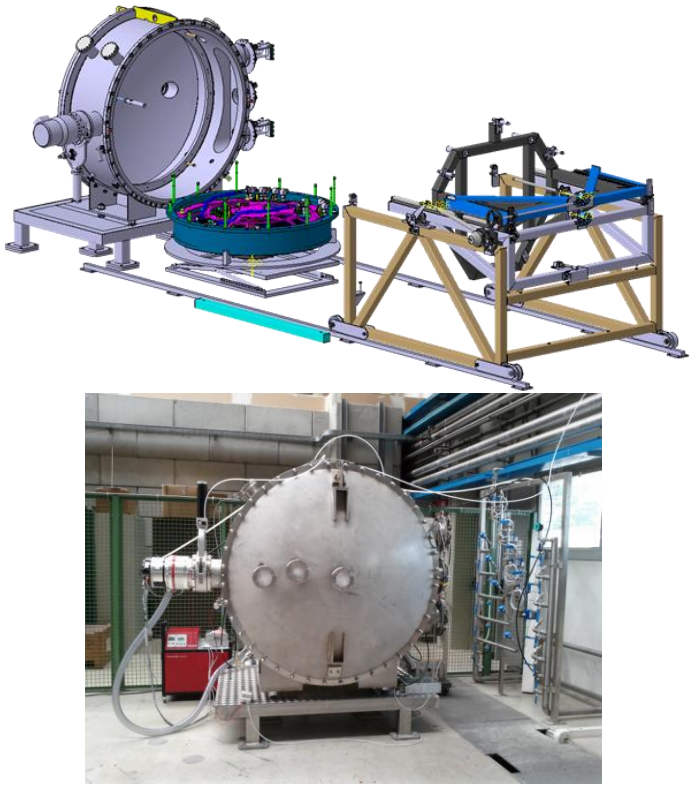

Figure 7. A sketch of the of the maintenance tool (left) and a picture of the production module housing the neutron converter module(right) .

The ball bearings sets for the driving motor/transmission shaft and the must be mandatory replaced.

The visual inspection of the converter, the water cooling, the delay window and the electrical components, is suggested at the end of each working period, at least during the first operative year.

The neutron converter has about 10.000 hours lifetime, this is more than 4 working periods, after which the wheel must be replaced.

The motor drivers are submitted to an integral dose about 10^6 Gy every working period and have to be replaced every year.

The graphite gaskets have to be replaced at every disassembling of the NCM.

The cooling panels, delay window and other mechanical components are submitted to a relatively low radiation rate and the accumulated damage is in the order of 1 dpa per 10.000 hours of operation. Their replacement may be planned after reaching the 5 dpa of damage.

Conclusions

The development's plan for the neutron converter is integrated in the general plan of development for the SPIRAL2 project. The development's plan is mainly based on the fulfilling of several actions related to the successful completion of the R&D program, the full design of the NCM, the manufacturing and testing of the NCM.

The manufacturing of the NCM and Delay Window are in progress and they delivery to Laboratori Nazionali di Legnaro is planned in October 2013.

The acceptance tests are planned to be performed within the end of the 2013 and the delivery of the full complex to GANIL/SPIRAL2 is foreseen within February 2014.

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6. The MYRRHA Project in Belgium

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MYRRHA, (Multi-purpose hYbrid Research Reactor for High-tech Applications), is a hybrid system, that combines a 600-MeV x 4-mA proton linear accelerator and a lead-bismuth eutectic (LBE) cooled fast spectrum research facility. MYRRHA can be operated in both sub-critical (accelerator-driven system (ADS)) and critical modes, allowing fuel developments for innovative reactor systems, material developments for GEN IV systems, material developments for fusion reactors, medical radioisotopes production and industrial applications (e.g. Si-doping). The system will also demonstrate the ADS full concept by coupling the three components (accelerator, spallation target and subcritical reactor) at a reasonable power level to allow operation feedback, scalable to an industrial demonstrator and allow the study of efficient transmutation of high-level nuclear waste.

Moreover, by branching off a small fraction of the proton beam, the ISOL@MYRRHA facility will be operated in parallel to the MYRRHA-ADS. This facility should use up to 200-□A x 600-MeV proton beam for the production of Radioactive Ion Beams (RIBs) via the Isotope Separator On Line (ISOL) method. By combining the high primary-beam intensity with selective ionization and a beam-purification system with high mass-resolving power, it will be possible to produce intense RIBs with high purity. ISOL@MYRRHA will be complementary to existing facilities, by focusing on experimental programs requiring long uninterrupted beam times. These are experiments which

- hunt for very rear phenomena,
- need high statistics,
- need many time-consuming systematic measurements,
- have inherent limited detection efficiency.

Measurements with high-intensity beams and extended/regular beam times are an important source of information for quasi all fields in science making use of RIBs, ranging from fundamental-interaction measurements with extremely high precision over systematic measurements for condensed-matter physics and production of medical radio-isotopes. In nuclear physics, e.g. determining precise values of extremely small decay branches (in the order of 10^{-6}) or crystal □-ray spectrometry with very high resolution can provide crucial experimental input for understanding aspects of nuclear structure. Long beam times could be also of interest for astro-physics, when nuclear reactions with small cross sections are involved, but the absence of a post-accelerator in the present design of ISOL@MYRRHA will prevent such kind of studies. Although higher-energy

secondary beams are not discarded for a later phase, only research with low-energy beams (up to 60 keV) is addressed for the moment.

Obviously, a strong synergy exists between the R&D and preparatory work for EURISOL and for MYRRHA (including ISOL@MYRRHA). Examples are developments for the linear accelerator, high-power target development, target-ion source systems for radioactive ion beam production, safety issues, detailed costing, dismantling plans, refining the RIB science case, building a RIB users community, etc.

7. Next-EBCB very fast charge breeding, production of fully stripped ions and cw beams with next generation Electron Beam Charge Breeder

A. Shornikov, F. Wenander - CERN

Motivation

Next generation Electron Beam Charge Breeders (Next-EBCB) can boost the performance of linear post-accelerators for fixed-target experiments as well as for injection into storage rings [1]. A Next-EBCB can be a unique source of very heavy ions (U/Th) in few-electron configurations and up to medium-heavy (Ba) in bare state. The main limitations of EBCB devices are their limited space-charge capacity and the pulsed ion extraction. Both can be improved in the next generation so that the breeder will not compromise the overall facility performance.

Although being complex and costly devices requiring some R&D, EBCB present a cost effective solution for the overall project. By decreasing the A/q ratio of the extracted ions one can reduce the length and therefore the cost of the linac. Next-EBCB can also be more effective in production of very low A/q for storage ring injections compared to foil stripping after the linac [1].

Several parameters should be improved simultaneously by an order of magnitude [2] compared to present EBCB several designs. The REXEBIS values are given within parenthesis:

- High electron beam energy up to 150 keV to ionize deep shells and produce lowest requested A/q (5kV);
- High electron-current density of about 104 A/cm2 so the breeding rate becomes compatible with the linac time structure, experiment data acquisition and lifetime of requested isotopes (100 A/cm2);
- High breeder capacity of about 1011 charges so all produced exotic isotopes can be loaded into the breeder without losses (109).

The performance of the Next-EBCB as well as existing breeders at ISOL facilities can be further improved by converting them into continuous highly charged ion-sources by means of intermediate trapping and debunching devices.

Apart from its direct impact on RIB/ ISOL facilities the R&D activities suggested in this project will also benefit other facilities such as injectors of highly charged stable isotope beams for space-related material research, biological experiments with heavy ions and solid state surface spectroscopy with highly charged ions.

Starting ground for the project

The main challenge for Next-EBCB is to produce the electron beam with requested energy, current and compression. Furthermore, one has to prove that ion injection and extraction from such an electron beam can be done efficiently. One needs to produce the electron beam, ensure it effectively captures 1+ ions, keep the ions while they undergo charge breeding, minimize the emittance growth and effectively extract them. It is important to mention that the actual electron beam parameters and machine physics studies can only be carried out with a fully equipped EBCB test stand, featuring a 5-6 T magnetic field and suitable for Brillouin-type electron guns, equipped with high voltage infrastructure and properly designed high-power electron collector.

Our proposal for Next-EBCB R&D is based on a unique combination. There is already an initiated program for the study of a High Energy Compression and Current (HEC2) electron gun by BNL and CERN, which takes advantage of an existing and fully functioning EBCB test stand at BNL. At the moment, June 2013, an important step in the HEC2 program has been reached as the first version of the HEC2 electron gun for short pulse test operation according to the BNL design [3] has been built at CERN. In summer 2013 it will be brought to BNL for tests at the BNL test stand.

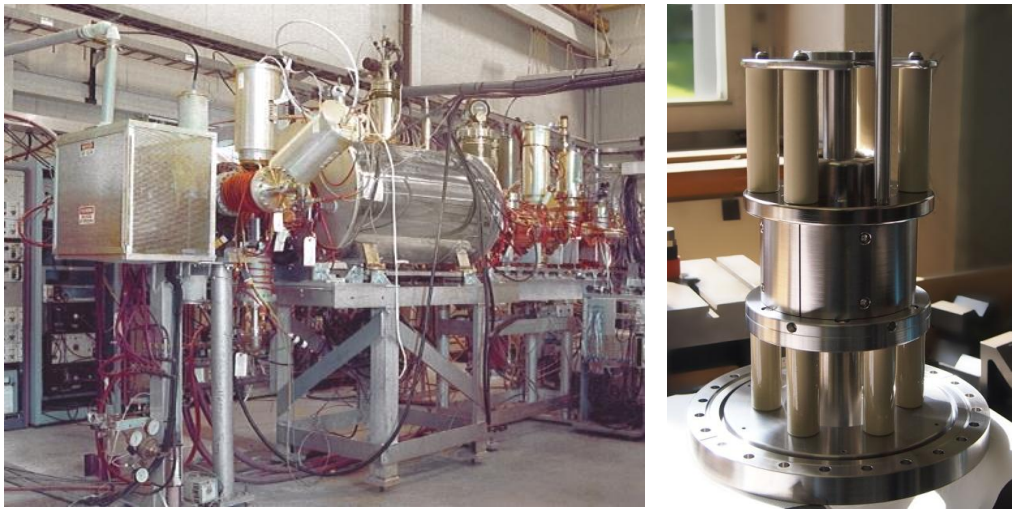

Figure 1 Left: BNL EBCB test stand. Suitable for HEC2 tests and operational. Right: HEC2 electron gun pre-assembled at CERN.

This test configuration shall provide proof-of-principle at energies up to 50 kV in short pulse operation mode.

R&D program

Operation at full voltage with long pulses and studies of the machine physics will require some changes to the gun design and the test stand (see Figure 2). The R&D to enable long pulsed and continuous electron beam operation includes improving the discharge resistance of the gun, modification of the electron repeller in the collector, and improving high-voltage and power ratings of isolation components and feedthroughs.

The modifications of the collector are motivated by the fact that HEC2 electron beam spreads in the collector area in a different way compared to a low compression beam of original TestEBIS design. The repeller electrode group should be modified to avoid damage of the optics when high power beam sweeps electrodes not connected to sufficient water cooling for a long period.

The machine physics R&D includes studies of interaction of the high compression beam with injected ions. The key questions are the breeder acceptance and the extracted ion beam emittance. To prevent emittance growth of the extracted beam and evaporative losses of the highest charge states, the implementation of in-breeder ion-ion cooling using low-charge state ions is a separate subtask for the machine development.

The first short-pulse test to be done using the BNL Test-EBIS with the entire gun chamber replaced by the new one.
Future cw tests will require some hardware development:

- Collector optics
- HW isolation
- Gun discharge receptivity
- Feedthrough power rating

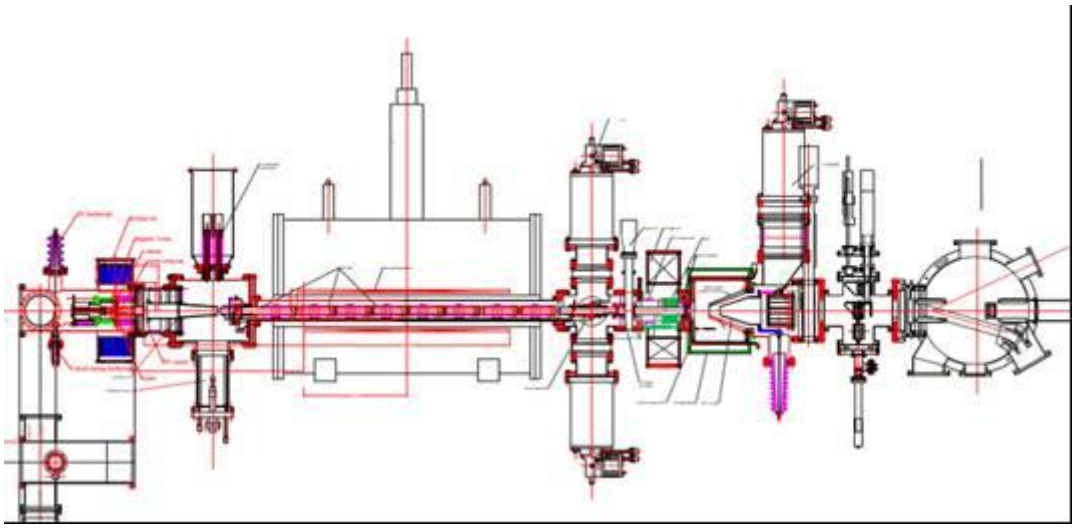

Figure 2. Hardware development and improvements for advanced machine physics studies

The most research intensive area is to suppress magnetron discharges at the gun, where they inevitable will start to glow if the voltage will be kept long enough in the presence of crossed electric and magnetic fields. The electron gun design developed by BNL is based on electron guns for high power RF tubes. In the RF tubes short pulsed operation with parameters close to requested for Next-EBCB was demonstrated. However, extinguishing the magnetron discharge is a new area. This requires careful 3D modelling (see Figure 3) as the axial symmetry shall be broken. After being numerically proven an updated discharge-free version of the gun shall be tested in long pulse/continuous operation mode.

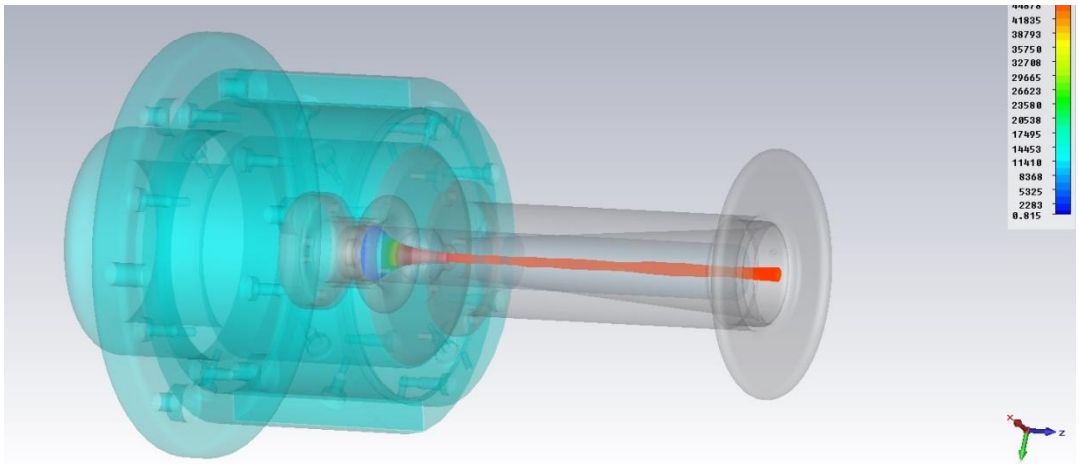

Figure 3. First test of full 3D space-charge limited electron beam with 49 keV energy and 10 A current using the CST software. The model is being integrated at CERN into CAD design workflow, has 2.74 M nodes, and the beam is created by 6170 individual trajectories. The simulation covers all aspects from emission to guiding magnetic field.

The current early phase of the program is entirely funded by the European side while BNL provides in-kind contribution in form of intellectual property (drawings, simulations, consulting), some components and access to the test infrastructure. Only limited manpower can be allocated on BNL side. The current financial support by the EU funds will end by October 2014. Therefore to continue R&D benefiting future ISOL facilities we request support in the framework of ENSAR-2 program.

Links to other ion sources projects and TNA in this JRA

In parallel with the HEC2 activities at CERN, there is an on-going project at GANIL aiming to use a debunching RF Paul trap for conversion of pulsed beam extracted beams from an EBCB into a quasi-continuous debunched beam (see contribution by P. Delahaye and E. Traykov elsewhere in this document). This can benefit both present and future ISOL facilities. Apart from its own project CERN could contribute to the debuncher project by operating the TWINEBIS – EBCB as a test ion source for the debuncher trap, if the additional required manpower will be covered by an external budget.

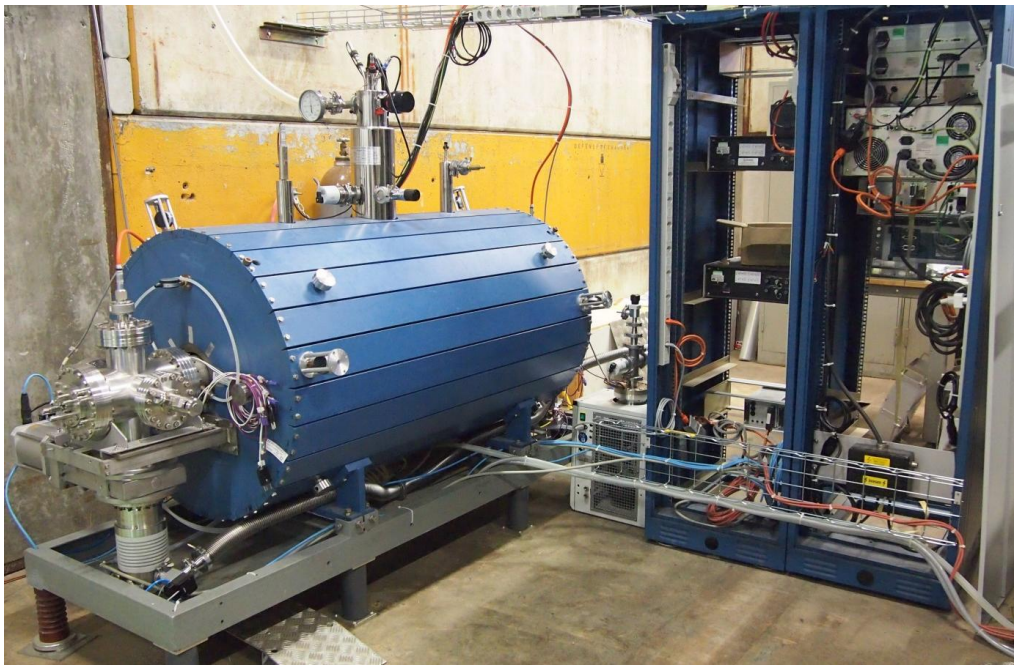

Figure 4. TWINEBIS is a replica of REXEBIS at CERN suitable as a test stand for the debunching trap and a test stand for intermediate upgrades of REX-like breeders. Presently getting operational after relocation from Manne Siegbahn Laboratory (Stockholm) to CERN.

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8. *Low-energy beam preparation, manipulation and spectroscopy - Summarizing the activities of PREMAS in ENSAR*

I.D. Moore, for the PREMAS collaboration

PREMAS is a Joint Research Activity (JRA) within the ENSAR Programme of the EU. The collaboration combines a range of expertise within the low-energy nuclear physics community to improve the beam preparation and manipulation of Radioactive Ion Beams (RIBs). Three key areas have been identified, each with a set of goals to be achieved over a period of 4 years: Novel RIB production techniques; advanced ion-beam manipulation, spectroscopic techniques and instrumentation; sources of pure and intense RIBs. These three areas provide much needed research and development, directly benefitting a number of EU Transnational Access facilities providing new opportunities to an ever-increasing user community.
The activities within PREMAS are divided into three main tasks, each coordinated by a specific institute. These tasks are further sub-divided into several sub-tasks overseen by additional institutes. Milestones must be attained throughout the 4 year period, and three final task reports serve as deliverables. The main tasks are summarized as follows:

Task 1: Novel radioactive ion-beam production techniques (KU Leuven)
The production of new RIBs, for example trans-lead nuclei, requires the development of selective and efficient laser ionization schemes, in some cases on-line when no stable isotopes exist. State-of-the-art target-ion source systems are now used at all large-scale facilities following a decade of detailed studies on atoms, molecules, photo-ions and surface ions at high temperature and in gas cells. New developments include tailored plasma conditions and gas-jet expansions. For example, by physically separating the thermalization and ionization zones in on-line gas cells the use of DC electrical fields can be applied to improve the ion beam purity. One of the most challenging obstacles at low-energy facilities is the contamination from isobaric reaction products and molecular contaminant ions, increasing as primary beam intensities are ramped up. A combination of laser selectivity and novel high-precision ion trap purification methods (for example for isomeric purification) is proposed as part of Task 1, resulting in ultra-pure RIBs for a variety of experiments.

Task 2: Advanced ion manipulation, spectroscopic techniques and instrumentation (JYFL)
The use of mass spectrometric techniques at low-energy RIB facilities enables a wide range of methods for the manipulation and preparation of short-lived nuclei prior to delivery to an experiment. Thus far, RF structures have been used for ion-beam cooling and preparation of well-defined ion bunches. Task 2 extends these applications to include novel methods of optical manipulation on stored ions in a RF cooler-buncher, to populate or deplete selective ionic states, to polarize the nucleus or even to ionize to a higher charge state. Penning-trap

technology has been widely used for precision studies. Within PREMAS, techniques are developed to optimize the coupling of ion traps to auxiliary instruments, diagnostics and novel detection setups to expand the applications of trap technology for nuclear structure studies.
Advanced spectroscopic techniques primarily focusses on applications of Resonance Ionization Spectroscopy (RIS) and its many variants, for example, in-source spectroscopy in hot cavities and gas cells, and exploratory studies of RIS within supersonic gas jets. The latter environment is highly attractive for spectroscopy due to the reduced atomic line-broadening mechanisms. In general, RIS has demonstrated impressive sensitivity for optical isotope shift studies, providing a wealth of ground state nuclear structure information (size, shape, spins and moments) of exotic nuclei. New developments in collinear resonance ionization spectroscopy (CRIS) are now pursued within PREMAS through the synchronous bunching of ion beams using an rf cooler-buncher, with low repetition rate pulsed tunable lasers.

Task 3: Sources of pure and intense radioactive ion beams (GANIL)
Ion beam purity is a major concern for nuclear physics experiments and thus a number of sub-tasks have been formulated to develop innovative techniques for the production of high purity radioactive ion beams by ionization in ECR sources and charge breeders. Although ECR sources have been successfully used for the production of pure beams of gaseous elements, the use of such sources as universal charge breeders have shown purity limitations due to background from the residual gas and support structures. Additionally, advanced methods are proposed for extending the number of possible beams, for example via the ionization of volatile molecules containing non-gaseous elements to produce pure beams of condensable elements such as carbon.

The coupling of ECR sources to targets is usually complicated by limitations in space, available power and connectivity of the target-ion source units. Activities within PREMAS are studying how to overcome these problems. In parallel, challenges exist in maintaining a high performance level after long irradiation times, an issue deemed critical for next-generation facilities such as SPIRAL-2 or HIE-ISOLDE for which the neutron flux will be significantly higher. Radiation-hard sources are thus being studied and novel magnetic field structures which do not require permanent magnets (which degrade after long term exposure to neutrons) are proposed, for example the ARC-ECRIS design from JYFL.

PREMAS – a selection of highlights
The development of laser ionization schemes using dye and/or Ti:sapphire laser technology is critical to the selective and efficient production of RIBs. Such work is often distributed among a number of laboratories and the data is then accumulated and shared within the laser community. In particular, the known excited atomic states of heavy elements are often limited to the first state in a possible 2- or 3-step ionization scheme. Indeed, even basic chemical

information such as the ionization potential (IP) may be missing completely. In a recent highlight, four PREMAS institutes were involved in a measurement of the first ionization potential of astatine, determined via laser ionization spectroscopy on a series of Rydberg states [1]. Astatine is the rarest naturally occurring element on earth. The new value serves as a benchmark for quantum chemistry calculations of the properties of astatine as well as for the theoretical prediction of the ionization potential of the heaviest homologue, super-heavy element 117.

Continuing with the theme of heavy elements, the reduced amount of experimental data in the upper part of the chart of nuclides is due to a lack of stable isotopes and low production cross sections. A search for an efficient ionization scheme has been performed for Ac (Z=89) at the University of Mainz, using the long-lived 227Ac isotope (t1/2=22 y). In addition a more precise value for the IP has been obtained [2]. One of the new schemes was subsequently utilized at LISOL, Louvain-la-Neuve, to ionize 212,213Ac isotopes produced in fusion-evaporation reactions and preliminary in-gas cell broadband laser spectroscopy was performed on 212Ac [3].

In order to perform high-resolution spectroscopy of high-Z elements, efforts are underway to address the limitation due to the laser bandwidth, which for solid state systems may be several GHz. Such intrinsic resolution severely hampers the ability to resolve hyperfine structure and thus to gain access to fundamental nuclear structure information. Similarly, the ability to produce pure isomeric beams is hindered. A relatively simple way to reduce the laser linewidth is by the addition of a second etalon into the cavity. At LISOL this has been demonstrated on the dye laser system and utilized on stable copper isotopes. Similarly, a dual-etalon system is now in operation for Ti:sapphire lasers at both JYFL (tested on copper) [4] and ISOLDE (in-source spectroscopy on Po, At and Au, and collinear RIS on Fr) [5]. Dramatic reductions in laser linewidth require more complicated laser developments, and members of PREMAS are constructing injection-locked laser resonators which have very recently demonstrated a linewidth reduction from ~5 GHz to ~10 MHz. This method was used in a successful study of the hyperfine structure of the atomic ground state of 229Th [6] and during 2013 in the determination of the hyperfine structure in 227Ac.

With the purpose of suppressing surface-ionized isobaric contaminations in RIBs produced with the hot cavity Resonance Ionization Laser Ion Source (RILIS), a Laser Ion Source Trap (LIST) has been developed by the University of Mainz. The LIST separates the regions of the laser ionization and the surface ionization through the use of a positively-charged repeller electrode. The first successful on-line test was performed in 2011 whereby Mg was laser ionized inside the LIST RFQ while surface-ionized Na and Al isotopes were suppressed by more than 3 to 4 orders of magnitude. In 2012, the first on-line physics experiment took place and laser ionization of Mg and Po was demonstrated [7]. In fact, the hyperfine structure and isotope shifts of 217Po and 219Po were measured for the first time using in-source laser spectroscopy due to the suppression of surface-ionized Fr by the LIST.

The main experimental conditions that prevent higher spectral resolution studies with a gas cell-based laser ion source could be overcome by performing the photo-ionization in the cold jet expanding out of the gas cell. Such a technique may be viewed as an extension of the aforementioned LIST method, in which neutral atoms are selectively ionized upon exit from the gas cell, within the gas jet, and are captured by the rf field of an rf multipole before transport to the mass separator. A key requirement is the need for a good geometrical overlap between the atoms and laser beams, and at JYFL initial work has been carried out to study the gas jet formation as a function of nozzle type [8]. On-going work at JYFL and LISOL has resulted in the first demonstration of spectroscopy on stable copper [9, 10] however a full implementation of in-gas jet laser spectroscopy for its use in the study of rare isotopes has not yet been accomplished.

The Collinear Resonance Ionization Spectroscopy (CRIS) beam line at CERN-ISOLDE is an experiment which combines the Doppler-free resolution of collinear laser spectroscopy with the high detection efficiency of resonant ionization. In an experimental campaign during 2012, the hyperfine structure of the isotopes 202-207,211,219-221,229,231Fr and of isomers in 202,204,206,218Fr was studied [11]. An experimental efficiency of 1% has currently been demonstrated and a non-resonant equivalent efficiency was determined to be <0.001%. For additional selectivity, RIBs containing several isomeric states may be sent to a decay spectroscopy station. In the latest developments, laser-assisted decay spectroscopy was performed on 204g,m1,m2Fr.

At GANIL, the use of an ECR charge breeder of the Phoenix type, together with a surface ionization or FEBIAD source, was found to be the most universal method for producing beams of condensable elements. The breeder is being installed at SPIRAL during 2013.

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9. R&D on radioactive ion sources at the TNA ALTO facility

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In the framework of European projects for future facilities and for the operation of ALTO facility, IPNO carries on various developments on ion sources to produce nuclear beams of interest. The works in the last two years have been achieved as planned in the previous EURISOLNET working group at CERN in 2011. These developments cover three major fields: plasma ion source, molecular beams and selective laser ionization. First, efforts have been invested to progress in the IRENA plasma ion source. Second, a new program has been achieved to carry out fluorination of Lanthanides. Third, an on-line installation for resonant ionization by laser has been made operational at the TNA ALTO facility. The status of these developments has been illustrated at this EURISOLNET working group at Jyväskylä.

IRENA ion source

During the EURISOL-DS, a plasma ion source has been designed to operate specifically in a high radiation environment such as the one considered in the MW target station. The tests achieved on the first designed prototype have validated the IRENA concept. In the previous EURISOLNET working group in 2011, tests on a second prototype were in progress. These tests could be achieved only at an extraction voltage at 20 KV due to various technical issues. After a few attempts to solve these issues, we decided to fully upgrade the extraction electrode system so as to address also the request of a new on-line electrode at ALTO for safety aspects. Anyway, all the planned tests at 20 KV have been achieved before changing the extraction electrode. The results obtained show that IRENA can be a very competitive radioactive plasma ion source, having already at 20 KV comparable ionization efficiencies for noble gases as the FEBIAD ion sources used on-line at the TNA ALTO facility.

The latest experimental results have been presented at the EURISOL Town Meeting at Lisbon in October 2012. Further information on the IRENA has been detailed in an article by S. Essabaa et al., recently accepted in Nucl. Instrum. and Method B.

The new extraction electrode has been designed with the engineering office of IPNO's Accelerators Division, and with the collaboration of ISOLDE-CERN: R. Catherall, S. Marzari and T. Stora. A major difficulty in the design of this new electrode comes from the limited volume of the electrode chamber which is much smaller than the one of the ISOLDE-CERN front-end. The design also has to integrate a selection of materials that withstand the radiation generated at ALTO by the impact of 10 µA electrons at 50 MeV on a thick target containing ~60 g of depleted Uranium. The new electrode has been assembled beginning of 2013 and successfully tested at the extraction voltage of 30 KV.

Meanwhile, based on the measurements obtained on the second prototype of IRENA, additional simulations have been carried out using the Lorentz-3EM code and the thermal code IDEAS to further optimize the second prototype.

R&D on the fluorination of Lanthanides

Experiments requiring neutron-rich lanthanide beams were accepted by the first ALTO PAC. To produce efficiently such beams at ALTO, the use of fluorination as a chemical process to get volatile lanthanide fluoride molecule has been considered. As data in the literature are heterogeneous, a development program has been initiated to study in a systematic way the various parameters involved in the production of lanthanide beams by fluorination. This program involves also collaborators from CSNSM: I. Deloncle, and the Tandar in Argentina: M.A. Cardona and D. Hojman. The main parameters to be investigated are the injected fluoride gas, the target temperature and the ion source parameters. The experiments to produce the molecular beams have achieved at the off-line isotope separator of the TNA ALTO facility. Taking account the various data in the literature, we decided to start the experiments with CF4 as the injected fluoride gas. CF4 has been preferred to SF6, although SF6 looks possible, mainly because SF6 is expected to be more corrosive at high temperature. Let's notice that SF6 is used at GANIL to produce stable beams of Ge but the configuration of the system is different from a standard high-temperature target-ion-source unit. We have used a tunable calibrated leak to inject the CF4 gas in the target volume. To best simulate off line the release of Lanthanides from standard UCX pellets which have a high amount of graphite, we have synthesized graphite pellets containing tracers of Lanthanides. The targets used for the first experiments have been obtained by mixing Lanthanide oxides and graphite powder. Then the pellets obtained by pressing the mixture have been reaction sintered at running temperature. SRD spectra have been achieved before heating, after heating and after experiment with CF4. The stable Lanthanides have been selected so as to observe no overlap. This is the reason why three distinct target series have been synthesized: the first series containing La, Pr, Eu and Ho, the second series containing La, Pr, Eu, Gd and Tb while the third series contains Ce, Pr, Sm, Eu and Y. The first experiments have been run with a standard FEBIAD plasma ion source. Systematic measurements have been achieved and among the main conclusions, one can notice that the yields of Lanthanide beams increase as a function of the CF4 flow and that the maximum flow of CF4 compatible with good running conditions is about 6 10-3 mbar.l/s. We have also observed that the yields depend very sensitively on the target temperature. The various systematic measurements achieved also highlight the reproducibility of the results obtained and a high correlation between the valence of the Lanthanide and the form in which the ion beam is observed: monatomic ion, mono-fluoride ion or di-fluoride ion. For instance, La, Pr and Ho have been predominantly observed as di-fluoride.

A second campaign of measurements has been achieved using a W ionizer to produce beams by surface ionization. Two target series have been tested: the first one containing Nd, Eu, Tb and

Yb while the second one contains Eu, Gd, Tb and Dy. The Lanthanide chosen covers a wide range of ionization potential and boiling point so as to well represent what the ion source can produce for all the lanthanides. The results obtained are attractive; in this case we could also observe Lanthanide beams. The same correlation between the valence of the lanthanide and the form of the ion beams produced has been observed. Through the systematic measurements, the reproducibility of the results has been globally observed. The few discrepancies observed could be related to some specific running of conditions. In particular the sensitive dependence on the target temperature has been confirmed. Although further tests are planned to better determine the optimum running conditions, the results obtained so far confirms the feasibility of such experiments on-line at the TNA ALTO facility, under efficient and safe conditions. In particular, after running more than three weeks with CF4, no chemical corrosion has been observed in the front-end and the pumping system.

Such a kind of systematic program is very attractive for all existing ISOL facilities insofar as the technique is very efficient to produce chemically reactive nuclei. This is the reason why a project proposal has been submitted to the scientific committee of ENSAR2. This proposal labeled BeamLab gathers CERN, GANIL, GSI, LNL-INFN and IPNO. Regarding the wide variety of chemical techniques and the improvable parameters, the developments considered in the BeamLab proposal will focused on the new nuclear beams which are currently the most attractive for the running TNA facilities. For instance, beam developments for nuclei in the group of transition metal are considered by adding partial pressure of dedicated chemical gas. The proposal consists in four interrelated tasks on target material and ion sources.

RIALTO: the laser installation for Resonant Ionization at ALTO

The RIALTO (Resonant Ionization at ALTO) is the on-line laser installation built at ALTO in 2011 to provide radioactive beams obtained selectively by a multistep photo-ionization. It has been validated in fall 2011 with the production of stable and radioactive Ga beams. The laser installation is on a shielded mezzanine above the ISOL separator and inside an air-conditioned hut at 21 °C. It has bee, achieved in close collaboration with CERN-ISOLDE. Beginning of 2012 RIALTO has been equipped with two new dye lasers from Radiant Dyes. Then we have achieved the ionization both of the ground state Ga and of the 826.2 cm-1 metastable Ga, using respectively the two-step schemes 287 nm + 532 nm and 294 nm + 532 nm. These works have been presented at the EURISOL Town Meeting at Lisbon in October 2012. The RIALTO now operates with an EdgeWave Nd:Yag pump laser which supplies pulsed 10 ns green beams at 532 nm with a frequency of 10 kHz, the power of this pump laser is 100 W. The two Radiant Dyes Narrowscan lasers cover the range of 540 -900 nm and are equipped with a BBO doubling unit to cover the range of 270 – 450 nm. Currently the RIALTO has been tuned to ionize Zn by the three-step scheme 214 nm + 636 nm + 532 nm (the 214 nm laser has been obtained by frequency tripling). These developments at RIALTO have been achieved with the collaboration of CERN-ISOLDE.

In order to study new ionization schemes and to check rapidly the existing ones, a reference cell has been designed by the engineer office of the Accelerators Division. The design has been achieved in collaboration with Mainz University. The Accelerators Division of IPNO is also involved in the study of laser ionization in a gas cell for S3 SPIRAL2; beginning of 2013 an ANR has been submitted to get national funds. The first workshop on the laser ionization at S3 is organized at IPNO, May 30th-31st.

10. Status of TIS R&D for SPES RIB facility

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SPES [1] is the acronym for "Selective Production of Exotic Species". The aim of this ISOL facility [2] is to provide high intensity and high-quality beams of neutron-rich nuclei to perform forefront research in nuclear structure, reaction dynamics and interdisciplinary fields like medical, biological and material sciences.
SPES represents an intermediate step toward the future generation European ISOL facility EURISOL[3]. The SPES project is supported by the Italian national laboratories LNL (Legnaro) and LNS (Catania). It is based on the ISOL method with an UCx Direct Target able to sustain a power of 10 kW.
The primary proton beam is delivered by a Cyclotron accelerator with an energy of more then 40 MeV and a beam current of 200 A. Neutron-rich radioactive ions will be produced by Uranium fission target [4] at an expected fission rate in the target of the order of 1013 fissions per second. The exotic isotopes will be re-accelerated by the ALPI superconducting LINAC [5] at energies of 10 AMeV and higher, for masses in the region of A=130 amu, with an expected rate on the secondary target of 108 pps.

THE SPES RIB FACILITY

The radioactive ions will be produced with the ISOL technique using the proton induced fission on a Direct Target of UCx [6] and subsequently reaccelerated using the PIAVE-ALPI accelerator complex [5]. A Uranium fission rate of 10^13 fission/s is foreseen. A Cyclotron with a maximum current of 0.750 mA rowing two exit ports will be used as proton driver accelerator with variable energy (30-70 MeV) [7].
Two proton beams can be operated at the same time sharing the total current of 0.750 mA. To reach a fission rate of 10^13 fission/s a proton beam current of 200 µA (40MeV) is needed; the second beam, up to 500 µA 70MeV, will be devoted to applications; mainly neutron production for material research and study of new isotopes for medical applications.
The ISOL technique for radioactive beam production is based on a driver accelerator which induces nuclear reactions inside a thick target. The reaction products are extracted from the target by thermal process, ionized and injected into a re-accelerator. In order to produce neutron-rich isotopes it is mandatory to perform fission reactions in Uranium or other actinide. The SPES choice is to use a proton beam to induce fission on a UCx target.

The driver is the proton cyclotron delivering beam on different targets. Two production ISOL targets are planned to be installed. The production target and the first mass selection element will be housed in a high radiation bunker. Before the High Resolution Mass Spectrometer (HRMS) a cryopanel will be installed to prevent the beam line to be contaminated by radioactive gasses and a RFQ cooler to reduce the input emittance of the HRMS. After passing through the HRMS, the selected isotopes will be stopped inside the Charge Breeder and extracted with increased charge (n+).

A final mass selector will be installed before reaching the PIAVE-ALPI accelerator, to clean the beam from the contaminations introduced by the Charge Breeder itself.

The injection to the ALPI Linac is based on the use of a new Radio Frequency Quadrupole, with the adiabatic bunching inside. In this way a high voltage platform can be avoided, and a higher overall transmission could be achieved.

The new RFQ will operate in a CW mode (100% duty factor) at a resonant frequency of 80MHz. This frequency is the same as that of the lowest energy ALPI superconducting structures. The injection energy of ions was set to 5.7 keV/u. This choice is a compromise between the desire to reduce the ion energy to simplify the LEBT and RFQ bunching section design and the need to increase the injection energy to reduce space charge effects. The extraction energy was set to 727 keV/u, higher than the output of PIAVE RFQ, to optimize the beam dynamics of the SRF linac.

THE SPES PRODUCTION TARGET COMPLEX

The most critical element of the SPES facility is the target unit, where the RIB is produced. It is composed of seven UCx disks, appropriately spaced in the axial direction, in order to dissipate the average power of 8 kW due to the proton beam passing through them. The discs are housed in a graphite box containing the UCx, with windows and dumps, both in graphite. The box is contained in a tantalum cylinder, which is resistively heated by passing a high current through it [8]. A tantalum transfer tube connects the target unit to the ion sources: in that system is able to plug several kinds of ion source devices[9].

The target chamber unit, made in aluminium, has a water-cooled base plate and is covered by another water-cooled bell jar. The chamber unit, about 40 kg. weight, is coupled to the proton and to the RIB beam lines, by means quick connectors. Two valves isolate the system respectively from the proton beam pipe and from the front end.

Since the target unit could be used several times, it will be shipped into a dedicated box with lead walls of 2 cm thickness.

The handling system must be designed in order to move the target chamber unit, housed in a lead container (700kg.), into the area where the target will be irradiated, open the container pull the target chamber and connect it to the beam lines. After the irradiation stage, the target

chamber must be, at the begin, removed from the beam lines, then stored in the lead container and finally send out of the “hot” area for further handling and long term storage.

CONCLUSION

The SPES project is one of the main nuclear physics developments in Italy for the next years. SPES is an up to date project in the field of nuclear physics and in particular in the field of RIBs; the project, now under construction, will represent an important step in the direction of the constitution of the European-project-facility EURISOL.
While the hardware installation is starting, the R&D program will continue and mainly focused on the target and ion source development and optimization. Paramount in this R&D stage is the creation of a wide network between the existing facilities around the world to share knowledge. This is the main activity that, with the help of EURISOL-NET has grown and hopefully it will continue to expand order to better use the huge amount of expertise available and which is needed in this kind of research.

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11. UCx developments at IPN-Orsay

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The Institut de Physique Nucléaire Orsay (IPNO) is strongly involved in the study of actinide targets for the production of neutron rich radioactive beams by the ISOL method (isotope separation on line) through active collaborations. IPNO has taken part in EURISOL-NET and is currently in charge of UCx target developments within the Spiral2 and the ENSAR ACTILAB projects. Moreover, for synthesis and physicochemical characterizations of the UCx samples (or chemical analogs), some efficient collaborations have been set up in France with Laboratoire de Sciences Chimiques (University of Rennes), Institut de Chimie Moléculaire et des Matériaux d'Orsay (University of Paris Sud) and Laboratoire des Solides Irradiés (Ecole Polytechnique).

The aim of the work pursued at IPNO is to increase the target density while improving the release properties. This could be achieved by establishing a compromise between uranium density (to stabilize UC instead of UC2) and the most open porosity while having less closed pores. First results, with the development of a method to measure the release properties of pellets after irradiation, were recently published[1]. Several techniques were performed in order to describe the structure (XRD) and microstructure (SEM, He pycnometry and Hg porosimetry) of the tested samples. These samples were synthesized from several routes (Table 1): arc-melting of uranium and graphite (ARC), carbureduction of uranium oxide (OXY and PARRNe) or oxalate (OXA and COMP30 like OXA in which 30 vol.% of graphite fibers are added). The densest samples made of UC are OXY and OXA whereas the porous ones are PARRNe and COMP30 which exhibit an excess of graphite. The release properties of thirteen elements have been determined by □-spectrometry measurements. The □ rays were collected from 2 pellets after irradiation, then one was heated at 1200°C or 1550°C for 1 h and the □-spectrometry was again performed on the 2 pellets. Clearly, the higher the temperature is, the higher the intensity of fission products (PFs) is important. At high temperature (1550°C), OXA sample provides the highest released fraction (Figure 1-a). However, the PFs releases for COMP30 and PARNNe are almost as high as OXA and theses samples exhibit particularly interesting releases at lower temperatures.

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Sample	Precursors	XRD analysis	*Open porosity (%)*	*Effective density ($g.cm^{-3}$)*
PARRNe	U_3O_8 + 6 C or UO_2 + 6 C	UC2 (+ UC+C)	60	8.5
OXY	UO_2 + 3 C	UC (+UC2)	40	13.5
OXA	$U(C_2O_4)_2,2H_2O$ + 3 C	UC (+UC2)	40	13.5
COMP30	$U(C_2O_4)_2,2H_2O$ + 3 C + 30 vol.% C fibers	UC2 (+UC+C+fibers)	60	10.5
ARC	U+C	UC	10	13

Table 1: physicochemical information of the 5 types of samples tested.

Following these promising early results, improvements were made to the protocol to perform new irradiation tests at ALTO facility in March 2013.

Concerning the synthesis, a planetary milling (1h at 325 rpm with 3 mm diameter balls in isopropanol) was performed on UO2 raw powder to reach a grain size of about 150 nm. Carbon nanotubes (CNT) were tested as a new source of graphite in pellets and a semi-automatic press was installed to perform uniaxial pressing and avoid any effect of the operator.

Concerning the heating, a new carburization bench was developed, working under secondary vacuum (instead of primary previously) and the temperature was calibrated by the melting of pure metals (purity > 99.9%) like V, Rh, Pt, Zr, Mo. A new design was realized for the heating of samples after irradiation allowing a fast cooling (10 min) with a good mechanical strength of the pellets.

About the irradiation tests, a new sample holder was used with a defined aperture to control the irradiated surface. The irradiation time was 20 min (10 min for GATCHINA, Table 2) and the waiting time was 30 min for safety. The analyses of the physicochemical characterizations and FPs release measurements are still in progress.

Four of the 7 kinds of samples are already analyzed. Synthesis methods and physicochemical information are summarized in Table 2. CNT samples (like PARNNE composition with CNT instead of graphite) were carburized at 1600°C for 20 min whereas 1750°C for 16h for the OXA and COMP30. Gatchina pellets were purchased and prepared by hot unixial pressing at high temperature. With an excess of graphite, UC2 is mainly stabilized (COMP30 and CNT) whereas a stoechiometric quantity of graphite (1 : 3 UO2/C) leads to UC even if a small quantity of sub-stoechiometric UC2 (UC1.86) is present. Again stabilizing the UC phase leads to a higher density. Moreover, on-line measurements, after a heating during 30 min (Figure 1-b), show that the Gatchina pellets released Sn but not Kr and Xe. Sn release is known to be controlled by effusion whereas Kr and Xe by diffusion. In consequence, the whole results show that the higher the porosity, the stronger the release of the diffusion-controlled elements is. The strange behavior of the release of Ru is not understood so far. However, these preliminary results show clearly

the diffusion in the pellet is correlated with the open porosity rather than with the effective density.

Sample	XRD semi-quantitative phase analysis (%)	*Open porosity (%)*	*Effective density (g.cm^{-3})*
OXA	76% UC + 24% UC_2	25	13.1
COMP30	4% UC + 81% UC_2 +15% CNT	48	10.1
CNT	12% UC + 88 % UC_2	59	8.5
Gatchina	82 % UC + 18% UC_2	5	12.2

Table 2: Summary of the synthesis of pellets made for the irradiation tests performed at ALTO in March 2013.

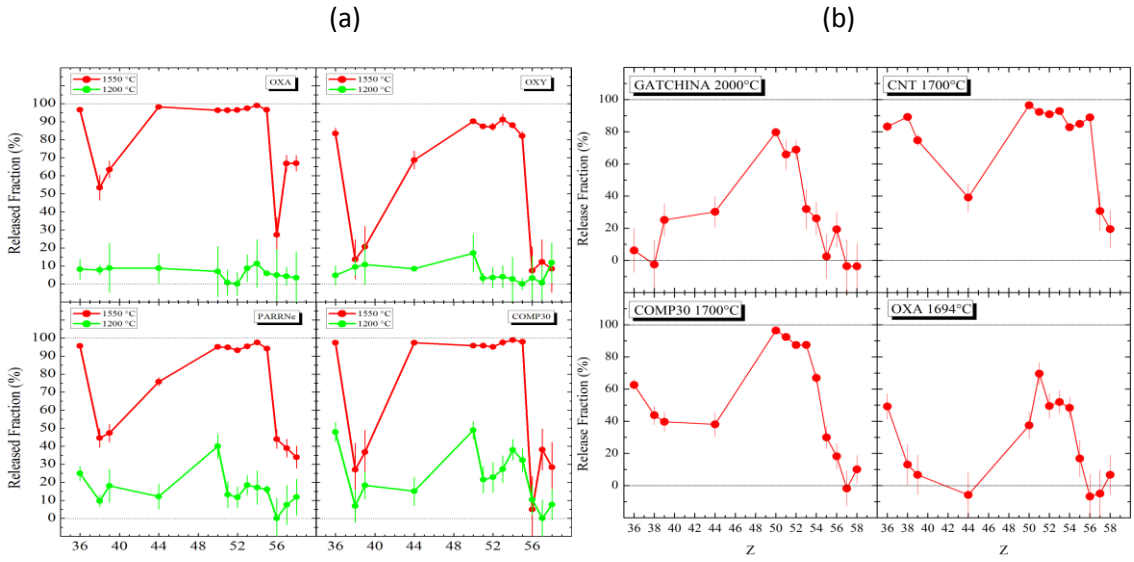

Figure 1: Release measurements for (a) preliminary tests to validate the method, (b) tests after improvements of the method.

12. μ-Mapping of Electronic and Structural Properties of Complex Uranium Compounds

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Keywords: uranium carbide, micro-XAS, non-irradiated fuels

Advanced materials based on uranium and thorium refractory compunds are a corner stone for large scale facilities delivering radioactive beams throughout the world. The technique, first pioneered at the Niels Bohr Institute in the 50's, exploits the interaction of an energetic particle beam with a thick target to induce nuclear reactions. The produced isotopes are stopped in the thick target, before they diffuse and are released, serving as a particle source for a secondary beam purified by mass-separation and delivered to experiments active in fundamental nuclear physics and astrophysics.[21] Along the years polyphasic porous uranium carbides became the reference material used in the different facilities across the world. It allows today accessing a wide range of radioisotope beams as for instance illustrated at ISOLDE, CERN.[1]
Uranium-based fuel materials constitute also the chore elements for nuclear electricity production, in which the neutron-induced fission reactions are used to generate the heat for further conversion into electricity. The function of the uranium-based materials is here radically different: the nuclear reaction products must be fully contained in the fuel rods to prevent any detrimental impact on the installations and the environment.[24] Specifically for the development of the next generation IV nuclear power plants, which aims at improving the safety and efficiency of nuclear energy production, active research on new uranium carbide materials is undertaken.[24]
In most nuclear power plants radioisotopes are produced as an undesired side-product of the exothermal fission process of uranium-235 under thermal neutron irradiation. Per contra, in radioactive beam production facilities, the radioisotopes are highly desired and are produced by sub-critical fission of uranium-238 induced by a high-energy neutron or proton driver.
Whereas an idealistic nuclear fuel detains all toxic radioisotopes the perfect ion beam production material releases all products of nuclear reactions instantly. Though antithetic, the material requirements in nuclear power and radioactive ion beam production are determined by the very same microscopic properties, which are accessible in a uniquely comprehensive way by the integrated techniques presented here.
On a fundamental basis, transport properties of actinide compounds, linked with complex localization patterns of the 5f electron shell, is an active field of research. Information of the chemical composition, phase and structure provides the necessary input to elaborate theoretical modeling.[7]

The need for a rational development of novel materials that display different transport properties calls for a careful characterization of their phase, chemical composition, and microstructure. However the intrinsic radiotoxicity of actinides makes this task challenging and thus requires dedicated infrastructures and techniques.[8]

For instance the path to provide a new generation of uranium based materials for particle production was long lacking this information. Only in the recent times has scarce data become available.[13] However a precise modeling of the diffusion and release properties of various isotopes can only be done knowing this information.[25] Indeed, recent data show that high isotope release rates are intimately related with the synthesis of novel type of nanostructures.[26]

We present here data on dense polycrystalline uranium monocarbide and porous polyphasic materials resolved at a submicron scale; this provides for the first time information on phase and porosity which directly influence the rate of release of the various chemical radioisotopes. Specifically extended X-ray adsorption fine structure (EXAFS), X-ray fluorescence (XRF) and micro X-ray diffraction (XRD) using micro-focused synchrotron radiation is reported here. The methodology was developed to prepare samples with high radiotoxocity and to bring them to the beamline X05LA at the Swiss Light Source (SLS), Paul-Scherrer-Institute (PSI), where these investigations took place. This opens up the possibility to extend these studies to a new generation of materials after irradiation, to observe the diffusion or accumulation of chemical species along their operation under energetic particle beam irradiation.

Uranium carbides of different stoichiometry, density and grain size are intensively discussed as future ISOL-type target materials, where currently a porous dicarbide with excess graphite is widely used. Recently proposed were sintered UC matrices of 5 µm to 20 µm grain sizes with densities close to the theoretical density.[9] The isotope production and release of this material was complementally tested at ISOLDE, CERN and IPNO, Orsay, France.[23]

Due to the complexity of these advanced, engineered materials a comprehensive knowledge of properties on the micrometric length scale is crucial to understand the global diffusion properties first of pristine and later under or after high energy proton irradiation. Therefore a series of microscopic electronic, chemical and structural investigations were performed on the concerned uranium-based materials. With an incidence exciting synchrotron beam close or at the uranium L_{III} absorption edge at 17.2 keV and a set of two X-ray detectors for energy or position dispersive detection respectively allow for simultaneous X-ray diffraction (XRD) powder pattern mapping and X-ray florescence spectroscopy (XFS). Returning with the 1.4 μm^2 synchrotron focus to a chosen sample position, extended X-ray absorption fine structure (EXAFS) measurements can be performed at a dedicated spot of interest, identified in the XFS or XRD maps.

So far a series of three non-irradiated materials were studied, clearly illustrating the power of the applied techniques to understand complex, multiphasic, structured radioactive materials. Samples were either prepared for measurement by producing a defined micrometric cross section using focused ion beam (FIB) milling in a scanning electron microscope (SEM) or they were investigated in their pristine macroscopic pellet shape.

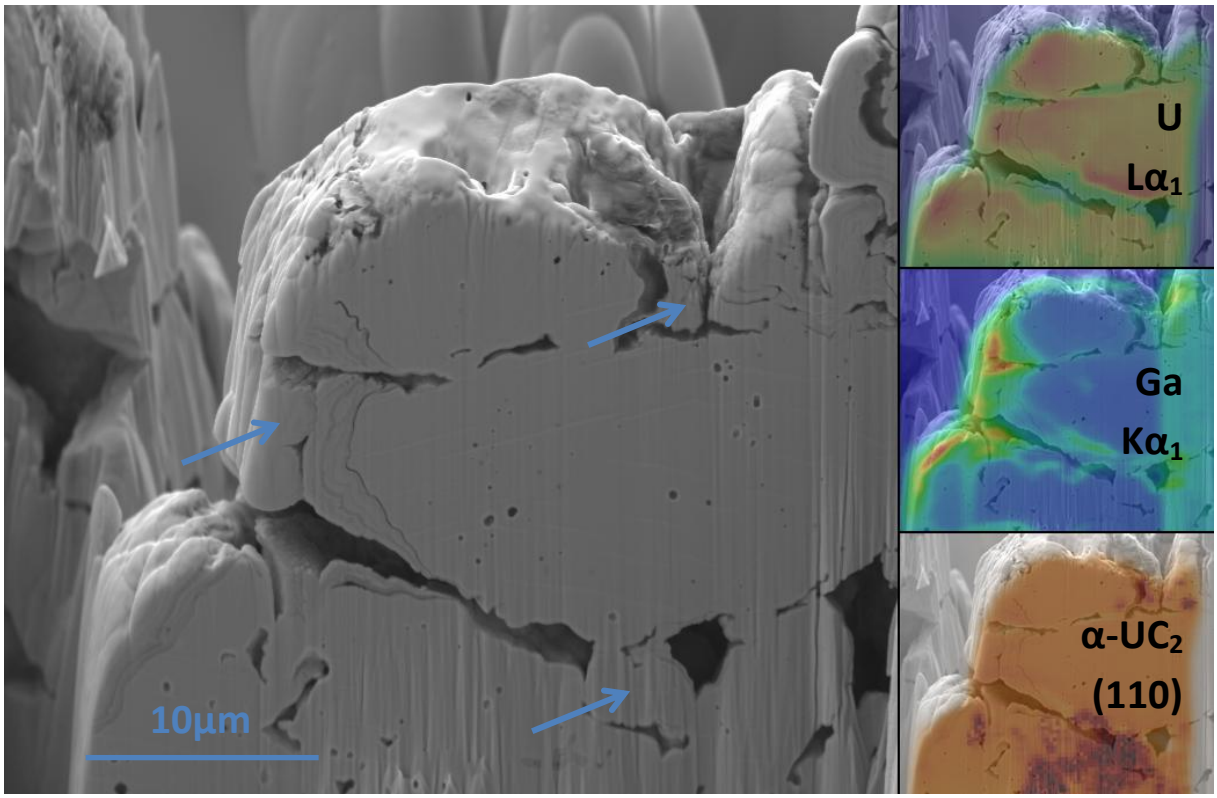

Figure 1. SEM image (electron energy 2kV) of a 7µm thick cross section through a uranium mixed carbide particle as prepared by the FIB technique (left). X-ray fluorescence emission map collected for the same particle in steps of 0.5µm at the U $L\alpha_1$ (top) and the Ga $K\alpha_1$ (middle) emission line. For Ga the false-color intensity scale is enhanced by 10^4 compared to the uranium image. XRD microscopy (bottom) showing the α-UC_2 (110) reflex intensity throughout the sample.

Porosity and microscopic distribution of both, chemical species and crystallographic phases are crucial parameter for most nuclear materials. **Fig.1** presents a series of sample images giving a broad overview over these properties:

From measurements of surface topography of these compounds the typical grain size was reported to be between 20µm and 50µm while bulk observations have never previously been performed.[6,13] Already the SEM image of a FIB thin cut reveals sub-particles of 1µm to 3µm, an observation that has major impact on the modeling of the release of fission fragments. A number of micro- and sub-micrometric pores can be identified; some of them appear to be closed as they are surrounded by material, others are connected to the surface of the particle. An observation, which could be made while stripping layer per layer was that these pores are in fact interconnected to a large extend, which results in a major fraction of the porosity to be open.

It is XFS, measured at the uranium U $L\alpha_1$ fluorescence transition that shines further light on the 3-dimensial structural arrangement. The upper right panel in Fig. 1 displays the XRF map of the same sample, showing the intensity of the uranium $L\alpha_1$ transition at 13.6 keV for each position of the

synchrotron beam spot on the sample. This intensity is proportional to the number of uranium atoms seen by the micro-focused synchrotron beam, the subshell photoionization cross section and the relative fluorescence yield. Therefore, this image directly represents the distribution of uranium density, hence pores, since the cut is believed to be made in a region of a single overall uranium carbide stoichiometric phase, where the C/U ratio is subject to small changes only. Comparing the SEM and the XFS map indicates that most pores are connected to each other and finally to the periphery of the uranium carbide particle and therefore being part of the material's open porosity.

During the FIB-based sample preparation a focused gallium beam was used leaving traces of Ga deposited on the particle's surface and the pore boundaries. The distribution of this contamination can be observed following the intensity of the Ga $K\alpha_1$ emission line throughout the surface. Microscopic XFS mapping is of significant importance when following the segregation or precipitation of fission fragments after irradiation, which will have major influence on particle diffusion and release properties. The intensity of the transition indicates sensitivity in the parts per billion regime, a concentration which is well within the production yield of most elements during the operation in either a nuclear power plant or a radioisotope production facility.

The electronic structure of compounds, where uranium occupies different valance states is of fundamental interest to understand the transition from localized to itinerant 5f electronic band structures.[7] While the L_{III} absorption energies are known for many uranium compounds, the edge energies for the respective carbides have yet only been reported recently for uranium monocarbide.[10,15] **Fig. 2** displays the EXAFS spectra measured on the uranium L_{III} absorption edge for the monocarbide (top) and the dicarbide (bottom). In both cases the synchrotron beam was focused to a size of $1\mu m^2$ to $3\mu m^2$ and the EXAFS spectra show strong oscillatory features, which indicates that the uranium atoms are located within a uniform lattice within this micrometric scale.

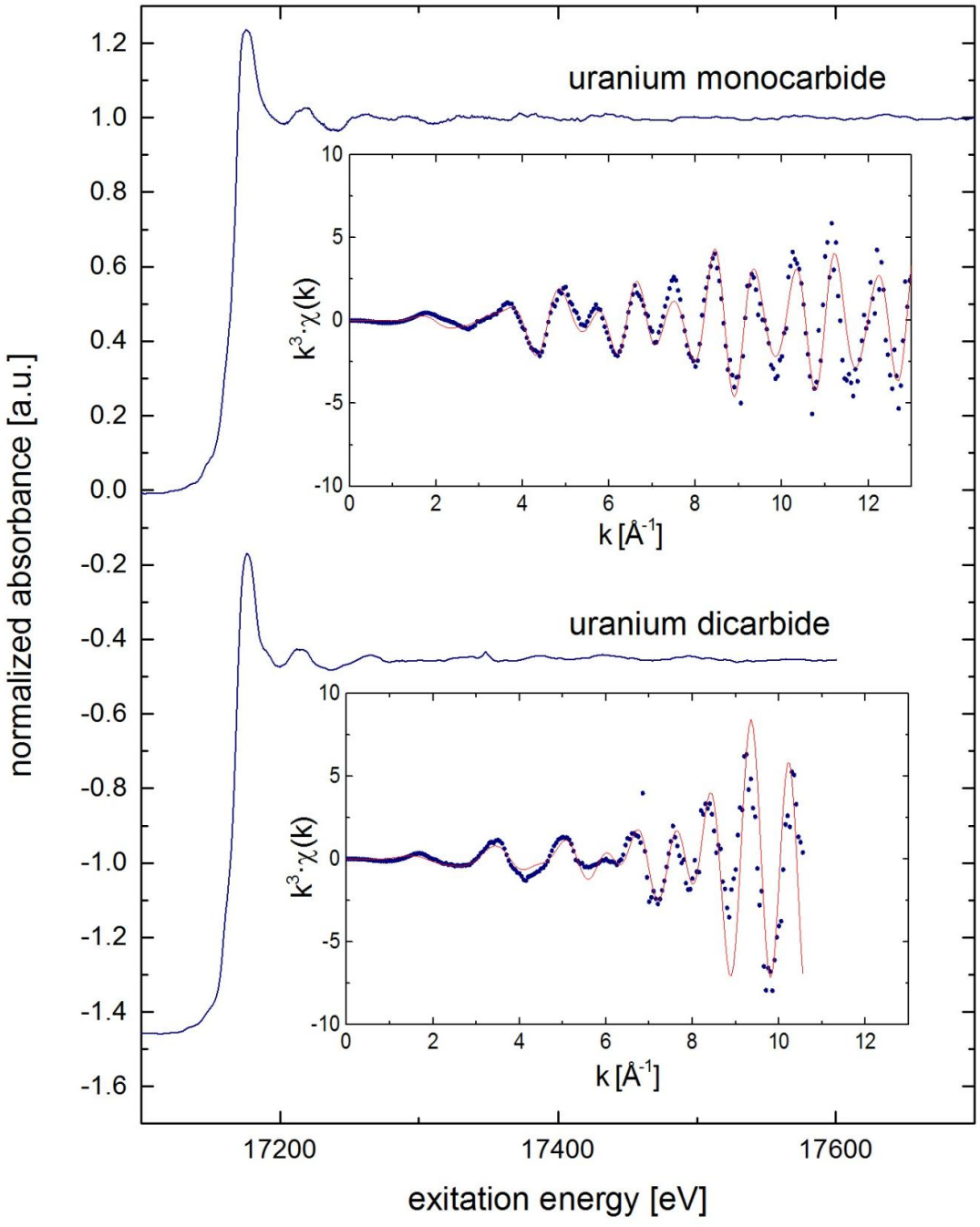

Figure 2. EXAFS spectra of uranium monocarbide measured with a 3µm^2 synchrotron focus (top) and uranium dicarbide measured with a 1.4µm^2 synchrotron focus (bottom). k^3-weighted absorbance as inset respectively (blue dots – data, red line – IFEFFIT fit for the respective structure as described in the text).

In order to extract the threshold energies the data was reduced including pre-edge background subtraction, followed by spline fitting and normalization. Using the Athena[10] code the position of the uranium L_{III} absorption edge was found to be E_0^{UC} = 17168.5(9) eV, in agreement with N. Vigier et al.[15] As expected from measurements and predictions for other U(IV) ions, the edge experiences a chemical shift towards higher energies for uranium dicarbide for which it is found here at E_0^{UC2} = 17170.0(6) eV.[16,17]
Both the UC and the UC_2 data were fitted using the IFEFFIT code after background subtraction.[19] The energy threshold shift ε_0 and the global amplitude factor S_0^2 where left open for the fit. For the EXAFS measurements sample positions were chosen from the XRD and XFS patterns comparatively where well defined structures were identified corresponding to space group Fm3m and I4/mmm for UC and UC_2 respectively. Further details for both fits are presented in the supporting material. The fits show good agreement with the lattices identified in the XRD patterns proving that EXAFS measurements can be exploited in the current setup to identify the electronic environment of prospective trace contaminations or fission fragments making use of their distinct sensitivity.

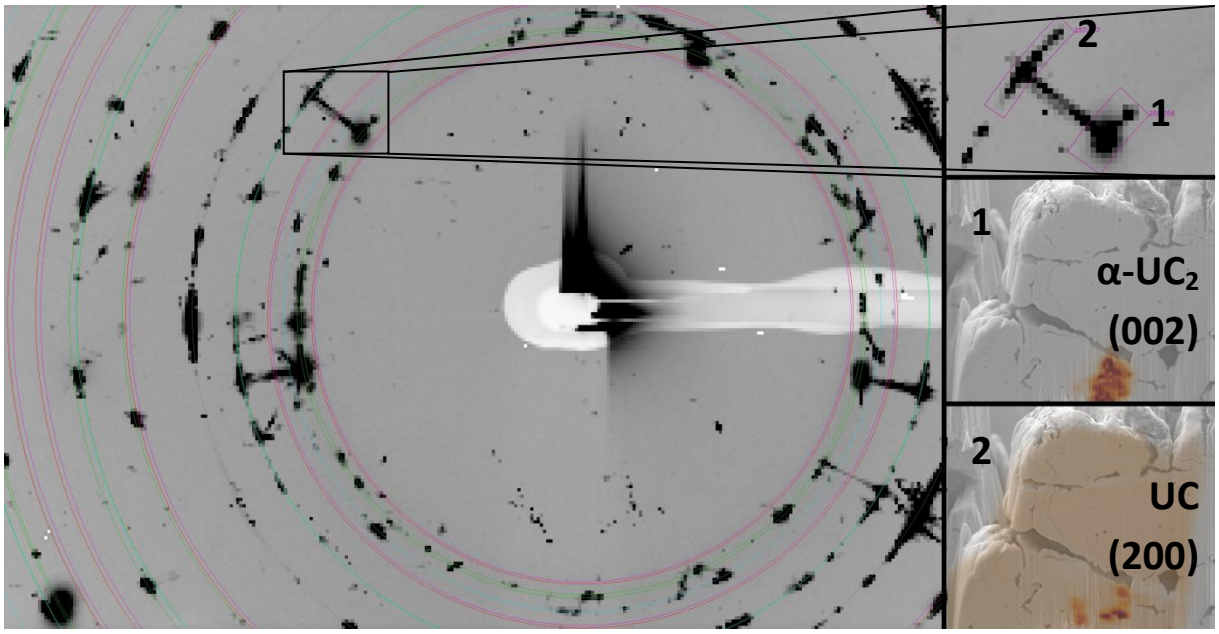

Figure 3. Image showing the superimposed XRD patterns of 10770 consecutive sample positions recorded during the full mapping (left). All major features can be assigned to low-index reflections of α-UC_2, β-UC_2, UC or UO_2 within the sample and additional Mo from the sample holder. A zoom on two reflexes (top right) reveals an inelastic scattering path originating from two coexisting sub-micrometric phase accumulations of α-UC_2 (middle) and UC (bottom) being stoichiometrically stabilized over a large range even at high temperatures.

µXRD can be used to reveal phase distributions in a micrometric scale but also 3-dimensional complex structural ordering phenomena at the same time. As shown in **Fig. 3** a wide range of phases can be identified and their distribution mapped. Single features in the pattern from Fig. 3

correspond to specific orientations within a certain crystallographic phase and high-symmetry reflection. All major reflexes could be assigned to either α-UC_2, β-UC_2, UC or UO_2. Additional molybdenum reflexes originate from the sample holder, where the sample was attached to by a carbon strip. A Rietveld fit after azimuthal integration (see supporting material) using the XRDUA code of the superimposed pattern exposes the contribution of each phase.[22] For the sample shown in Fig. 3 this is mainly α-UC_2 (76%), followed by UC (17%) and β-UC_2 (≈2%).
Besides this detailed insight into the microscopic phase distribution, additional characteristics can be identified. Especially notable are the tails along Δq or 2ϑ connecting two or more reflexes of different phases caused by inelastic scattering (e.g. as seen in Fig. 3 top right). Uranium dicarbide is known to be a material of high stability during high energy proton irradiation at high temperatures of more than 2300K. However it was not known, where this outstanding stability arises from. Following the spatial distribution of these features provides an explanation for the high thermal stability. UC_2 and UC are coexisting in sub-micrometric crystallites over a range of several micrometers due to stoichiometric imperfections favoring one of the two compounds. Sintering is therefore restricted until additional carbon defuses into this zone and transforms the mono- to the dicarbide or until the dicarbide decomposes to monocarbide and graphite. All these reactions are thermodynamically unfavorable even at elevated temperatures, causing the indirect structural stabilization.[19]
In a second sample investigated from the same batch but with a different thermal history these tails of inelastic scattering are nearly absent. In this sample the fraction of cubic UC derived from the Rietveld fit is well below 1%.

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