ECOS (European Collaboration On Stable ion beams)

ENSAR NA Report

1. The ENSAR ECOS Network Activity

The ECOS (European COllaboration on Stable ion beams) working group has been appointed by NuPECC in 2004 with the following tasks:

- Describe and access the research perspectives with high intensity stable-ion beams.
- Categorise existing facilities and their possible upgrades.
- Identify the opportunities and specifications for a dedicated new facility in Europe.

The ECOS working group has prepared a report which is available at the NuPECC webpages and which has been published in 2007. One of the important recommendations of the ECOS working group is to ensure a strong support from both the nuclear physics community and the funding agencies for existing stable-ion-beam facilities not only for their accelerator-system development but also for the instrumentation and experimental infrastructure needed to host dedicated research programs. The other important recommendation is that a new dedicated high-intensity stable-ion-beam facility in Europe, with energies at and above the Coulomb barrier, is considered to be one of the important issues to be discussed in the Long-Range Plan of the nuclear physics community. The objectives of the proposed ECOS-Network are related to these two recommendations and they are twofold:

i. Bring together and coordinate the expertise that is available in the European countries in order to achieve the research and development activities in essential aspects related to the production and use of high-intensity heavy-ion beams (Task 1). The important aspect related to the development of high-power ion sources is the objective of JRA01-ARES with which the NA02-ECOS will have a significant synergy.
ii. Optimise resources and manpower for the upgrade and development of various stable-ion-beam facilities in Europe in order to optimize their scientific output (Tasks 2 and 3). From this point of view, NA02-ECOS has a direct link to the TNAs delivering stable ion beams to the users community in Europe. These are TNA01-GANIL, TNA02-GSI, TNA03-INFN, TNA04-JYFL and TNA05-RUG. In order to achieve its goals, NA02-ECOS has been broken down into 4 tasks:

Task 1: *High power thin-target technology (participants: IN2P3 + GANIL+GSI)*

The maximum usable primary beam current with thin targets is among others determined by the long-term stability of the thin targets under irradiation. High beam intensities lead to a considerable heating of the targets, and, hence to thermal stress, possibly phase transitions, oxidation or reduction of the chemical compounds and diffusion into the target backing respectively. We propose to study these phenomena in detail and to compare for example the performance of thin actinide targets as function of the production method (painting, spray-painting, electrolysis, electrode position, evaporation and sputtering), the used chemical compounds (oxide, carbide, others) and backings/coatings respectively. The way is to bring together labs that use different techniques for target preparation and those that can test the target performance under real conditions. For this task ECOS will have the duty to organize the collaboration and exchange of expertise on the development of high power target technology.

Task 2: *Synergies in Superheavy Element Research (participant: GSI + GANIL+JYFL)*

The study of Superheavy Elements (SHE) is one of today's most challenging interdisciplinary research fields. It brings together nuclear physics, atomic physics, chemistry and theoretical physics. Over the last years researchers from the different disciplines have continued to strengthen exchange of ideas. The ECOS community proposes to use this Network in order to enhance synergies among the research groups on a European scale. For this task ECOS is aiming for bringing together the groups with research activities on SHE using high intensity ion beams for an exchange of new ideas and techniques related to the use of very high intensity stable beams.

Task 3: *Organization of bi-annual ECOS Workshops*

In order to optimize resources, two workshops will be organized with parallel sessions dedicated to all aspects of the technical developments and research activities using stable ions beam facilities in Europe. The second workshop will be coupled to the NA town meeting.

Task 4: *Coordination of stable ion beam facilities in Europe and organization of the network*

In order to achieve the goals of the ECOS NA and to foster synergies, collaboration and scientific exchange, a number of meetings have been organized (task 3). This report on task 2 is mainly based on the strategy discussion and in its results performed during the FUSHE 2012 workshop and follow up activities initiated by it. The FUSHE 2012 workshop was held in Weilrod, Germany, from May 13th to 16th 2012 covered all subjects related to superheavy element research including experimental, instrumental and theoretical questions (http://www.ensarfp7.eu/projects/ecos/workshops-meetings/fushe2012). The workshop was attended by about 90 participants from all institutions involved in SHE research worldwide. It was organized in 7 sessions with a strong emphasis on discussions including Theory, Experiment and Instrumentation in an integrated fashion in order to foster a constructive dialogue between theory and experiment. The sessions were organized as a combination of invited talks followed by a topical discussion for the subjects:

- SHE Synthesis
- Nuclear Structure of SHE
- Chemistry
- Atomic Physics and Alternative Approaches,

Covering all three disciplines

- Theory

- Experiment
- Instrumentation.

A document reporting on the findings of this workshop is in preparation and it is briefly summarized in this report. It covers all aspects aimed at by task 1, and will be published in an international peer reviewed scientific journal. This document is being prepared by an international writing group:

D. Ackermann[1], E. Litvinova[2], D. Boilley[3,18], Ch. Stodel[3], B. Avez, W. Barth[1,5], M. Block[1,5], Ch. Düllmann[1,5,6], S. Fritzsche[7], P. Greenlees[8], R.-D. Herzberg[9], F.P. Heßberger[1,5], W. Loveland[10], W. Nazarewicz[11], V. Pershina[1], G. Pollarolo[12], J.Roberto[13], P. Schwerdtfeger[14], A.Türler[15,16], J. Uusitalo[8], D. Vretenar[17]

[1] GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany
[2] Western Michigan University, Kalamazoo, U.S.A.
[3] GANIL, Caen, France
[5] HIM, Mainz, Germany
[6] University of Mainz, Mainz, Germany
[7] University of Jena, Jena, Germany
[8] University of Jyväskylä, Jyväskylä, Finland
[9] University of Liverpool, Liverpool, U.K.
[10] Oregon State University, Corvallis, U.S.A.
[11] Michigan State University, East-Lansing, U.S.A.
[12] Universit di Torino and INFN, Torino, Italy
[13] ORNL, Oak Ridge, U.S.A.
[14] CTCP Massey University, Auckland, New Zealand
[15] University of Bern, Bern, Switzerland
[16] PSI, Villigen, Switzerland
[17] University of Zagreb, Zagreb, Croatia
[18] Normandie Université, Caen, France

Concerning task 1 a dedicated conference was held during the funding period of the ECOS-NA with the 26th World Conference of the International Nuclear Target Development Society (INTDS 2012) from August 19th to 24th, 2012 at the conference center Erbacher Hof in Mainz, Germany (http://www.intds.org/).

Task3 resulted from a number of meeting and workshop that took place during the funding period of ECOS-NA and in which many the steering committee has played a major role: D. Ackermann (GSI), F. Azaiez (Orsay, Chairman), G. De Angelis (LNL), M. Lewitowicz (GANIL), A. Maj (Krakow), I. Martel (Huelva) and R. Julin (Jyvaskyla).

1. The deliverables:

In the following the three reports corresponding to the three deliverables of the ECOS Network Activities are given:

D-NA02-1: Report on the development of high power thin-target technology with special emphasis on new techniques and methods that will allow increasing the primary beam intensity usable with such targets.

D-NA02-2: Report on the research activities related to SHEs and on the achievement made in this research field

D-NA02-3: Report on the collaborations and synergies between facilities providing stable ion beam facilities in Europe initiated and driven by ECOS network

***D-NA02-1:** Report on the development of high power thin-target technology*

In the study of superheavy nuclei, a major experimental concern is the behavior of targets under highly intense heavy ion beam irradiation as well as the manufacturing of targets and the availability of target materials.

The material supply is particularly problematic for actinide targets because of the scarcity and the radioactive properties of the starting material. Indeed actinides are produced in nuclear reactors and often require sophisticated facilities and equipment for producing and separating the required isotopes in sufficient quantities. Some actinides can only be produced in national facilities situated in the U.S.A. or Russia, and the full cost of production and separation is very high. Assuming the actinide material is available the transformation into thin foils of appropriate thickness and quality is also challenging. Due to the high activities involved, the development of actinide targets requires specific knowledge of radiochemistry and the material can only be handled, transformed and transported by laboratories with special licenses granted by governmental authorities.

The behavior of the target foils under irradiation is related to the properties of the selected heavy ion beams and the structure of the target foils. On one hand, the properties of the beam and the spectrometer (such as energy, intensity, spatial dimensions, acceptance and transmission) constrain the thickness of target foils to be only in the order of some few hundreds of μg/cm^2. On the other hand, thin backings are required for state of the art fabrication techniques and the prevention of undesirable beam scattering. These prerequisites result in multiple risks for the targets upon irradiation:

- heating of the foil due to the beam power loss
- atomic and chemical reactions due to the beam impact at the material interface between backings and targets
- mechanical stress and radiation damage.

These phenomena of material transformation under irradiation conditions must be carefully controlled during the use of these targets in order to ensure safety and target survivability.

Her we review present and future target stations used for superheavy element studies, with a focus on target technology including material supply, fabrication techniques, and target characteristics. Some perspectives for improved target technology are also presented.

1. Target stations and its environment

Over the last two decades, beam intensities for the study of superheavy nuclei have ranged up to a maximum of a few particle μA at Coulomb barrier energies (5-7MeV/A) for target thicknesses of the order of $\approx$ 300-500 μg/cm^2. Stable material like lead or bismuth was evaporated on a carbon backing with a thickness of $\simeq$ 30-50 μg/cm^2 and covered with a carbon layer of $\doteq$5 μg/cm^2 to prevent sputtering. For radioactive targets, actinides were electrochemically deposited on rolled titanium foils of about 2 μm thickness. These conditions imply a deposited power in the material layer from a few to tens of Watts that needs to be spread over large surfaces in order to prevent overheating and eventually melting of target. The current targets stations for super-heavy research consist of fast rotating wheels on which the targets frames are mounted.

With planned upgrades at Dubna, GSI and GANIL, beam intensities are expected to reach tens of particle μA, which implies the necessity for larger wheels and/or higher velocities and/or cooling of the targets in order to avoid thermal destruction of the targets upon irradiation, an example is given in for the S^3 project.

In order to control the integrity of the target material, the target stations are equipped with various detectors and probes to prevent or detect damage and to trigger

beam stoppers.

Typical instrumentation comprises for example:

i. Rutherford detectors placed behind the foils at 30° or 45° with respect to the bema direction to measure the energy and yield of scattered beam-like and target-like particles at low beam currents.
ii. Electron guns to monitor the “transparency” of the targets under irradiation by exploiting the thickness dependence of the angular straggling of the electrons passing the foils. Thanks to the methods spatial sensitivity even pin holes can be detected.
iii.

2. Thin foils (stable and actinide material and strippers)

In the studies of super-heavy elements, production targets are required to be in the order of 500 μg/cm^2 with a surface homogeneity of $\pm$ 2% over an active target area varying from 3 to 15cm^2. Techniques of target fabrication like thermal evaporation or magnetron sputtering are suited for stable targets because of their high yield. The yield is an important criterion for highly isotopically enriched material that could be very expensive. For actinide targets, the techniques differ according to the availability of the material. For ^{238}U-targets magnetron sputtering is an option since it is neither highly radioactive nor very expensive and metallic sputtering electrodes are commercially available. For highly radioactive and sometimes very scarce actinides electro-chemical methods like molecular plating with much higher yields and the possibility of recovering most of the material after the process is mandatory.

The main requirements for superheavy element targets and their fabrication process are:

- High chemical purity of the material prior to deposition in order to reduce the background signals in the detection system from unwanted reaction products.

- Recovery of the used material because of its scarcity and price.
- A small and simple process set-up
- High deposition yield

The fabrication processes imply that the material has to be deposited on backings, such as titanium foils of $\approx 2\,\mu$m thickness for actinides, and carbon foils ($30\,\mu g/cm^2$) for lead or bismut. In order to prevent appreciable sputtering of material because of beam irradiation, stable targets are covered with carbon by fast evaporation techniques. For the production of actinide targets alternatively also painting is still used in selected cases to obtain thin layers with the required thickness. However, targets produced by this technique may suffer from poor stability of the layer in long-term irradiation cycles. Sputtering can be used to produce layers of metallic uranium (^{238}U) on Ti or carbon backings. This method is limited by the availability of a (metallic) sputter-target. Taking into account the previously mentioned limitations molecular plating is currently the only production method for actinide targets in cases where the desired actinide material is available in very limited amounts or possesses a high specific activity.

For stable targets, the major issue for target durability is the heating of the material due to the beam power losses deposited during irradiation; the melting temperature is then a critical point. Applying chemical compounds that have higher melting points than the pure metals, can be very useful. For example, for lead and bismuth, commonly used for the synthesis of SHEs, with melting points of 328℃ and 271℃, respectively, metallic targets have been successfully replaced by the compounds PbS and Bi_2O_3 with melting points of 1116.85 °C and 824 °C, respectively. In order to enhance the target lifetime, the GSI group has implemented further improvements in their target systems, such as enlarging the irradiated target area to reduce the thermal stress on the material. In addition target cooling as well as the transformation of the beam intensity distribution from the typical Gaussian shape to an almost rectangular distribution by an adequate ion-optical treatment of the beam have been investigated.

3. Material supply for rare actinides

Much of the recent progress in SHE research has been accomplished using actinide targets in combination with intense beams of the neutron-rich ^{48}Ca. With this so called “hot fusion” technique, pioneered and implemented at Flerov Laboratory of Nuclear Research (FLNR) at the Joint Institute for Nuclear Research (JINR) in Russia, decay data assigned to six new elements and more than 40 new isotopes with atomic numbers 113-118 have been collected in the past decade. The actinide targets have included ^{238}U, ^{237}Np, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, ^{249}Bk, and ^{249}Cf. Many of these target materials were provided by the Oak Ridge National Laboratory (ORNL) as part of the Department of Energy (DOE) Office of Nuclear Physics Isotope Development and Production for Research and Applications Program.

This program maintains inventories of many actinides, including isotopes of Pu, Am, Cm, and Cf, as well as production facilities for short-lived Cf and Bk, and chemical separation and purification facilities for all actinides. This infrastructure includes the High Flux Isotope Reactor (HFIR) and the Radiochemical Engineering Development Center (REDC), a unique complex of a world-class research reactor for intense neutron irradiation, heavily-shielded hot cells for chemical processing and separations, and radiochemical labs and glove boxes for purification and analysis.

Most recently, the HFIR/REDC complex was used to produce ^{249}Bk for element 117 experiments at JINR and element 117 and 119 experiments at GSI. Nearly 50mg of Bk were produced in two year-long campaigns that included neutron irradiation of Am/Cm seed material in HFIR and chemical separation and purification at REDC. The processing, which is similar for most actinides, includes multiple dissolution, extraction, ion exchange, and precipitation steps and results in material that is chemically pure. This material can then be electrodeposited on thin films, typically titanium foils, for use as ion beam targets in super-heavy element experiments The available paths to gain in production yield of berkelium targets include irradiation of pure ^{248}Cm instead of mixtures of the 244 and 248 isotopes, and the use of thermal neutron filtering. Assuming a perfect Cd cutoff of the thermal neutron flux and target material consisting of 1.0 g of ^{248}Cm (97%) and a HFIR cycle time of 23 days, with seven HFIR cycles, more than 80 mg ^{249}Bk is achievable. Along with the ^{249}Bk production the isotope ^{251}Cf (9 mg) is also produced. Higher production is possible with increased Cm feedstock. For ^{251}Cf targets, the project EMIS (ElectroMagnetic Isotope Separator) at ORNL, configured as an actinide separator located in a hot cell, would enable its separation with a rate of 1mg/h. The useable yield of the isotope ^{254}Es is estimated to be 40 μg with irradiations of 1.0 g of ^{252}Cf.

4. Production techniques

The production technique used for most of the actinide targets is electrochemical deposition by molecular plating. Its main advantages are the very efficient deposition yield, up to 90% and the “simple” procedure with one single deposition step for thicknesses of 500-1000 μg/cm^2. As the actinide material has to be deposited on a backing, a precise study for the backing material most suitable was done at GSI. It resulted in the application of about 2 μm titanium foils produced by cold rolling and characterized in respect to purity, absence of pinholes, thickness homogeneity better than ±2% and good behavior under high beam intensities.

Some alternative target production techniques are considered for materials other than actinides. For instance, the polymer-assisted deposition (PAD) is used to create crack-free homogeneous metal oxide films of europium, thulium and hafnium. The method consists of mixing a metal-oxide with a polymer solution; the metal-organic films are formed by spin coating the silicon substrate with this solution. Target thicknesses up to 600 μg/cm^2 are possible. It would be worth testing this method for actinide elements and their behavior under irradiation.

Some other alternative described are the electro-deposition using ionic liquids, the use of super-hydrophobic surfaces or of inter-metallic targets. In the first method ionic organic salts, liquid at room temperature, serve as solvent for metal ions. With backings pretreated in a way that a super-hydrophobic surface is formed, improved targets with a more homogeneous deposition of metal-oxide/nitrate from aqueous solution by simple evaporation of single drops are achievable. This technique has been used successfully for targets in a laser ablation ion source. The use of inter-metallic targets has also been considered, applying molecular plating of a lanthanide or actinide compound on a palladium backing which is reduced by heating it in a hydrogen atmosphere. First in-beam tests were performed.

5. Characterization

Characterization of targets is of importance concerning physical parameters (layer thickness and homogeneity) as well as for understanding the process of deposition (yield). The standard target characterization techniques include:

- Direct radiometric determination of the target thickness by α-particle- or γ-spectroscopy.
- Yield determination using neutron activation analysis of the supernatant solution subsequent to deposition
- radiographic imaging to check homogeneity of the deposited actinide layer.

6. Perspectives

In the coming years, most facilities will work on an upgrade of their accelerator in order to gain a factor of 5 to 10 for high intensity beams. Research and development in the target material, production techniques, handling and control therefore are major issues. There is a need to explore the limits of the current target technology, to search for alternative backing materials, to develop new methods for production and to study the interaction of target material with backing under long irradiation conditions with high intensity beams. Results from these studies have to be tested in realistic beam times with high intensity beams. In parallel to the target fabrication, target characterization (pre- and post-irradiation) with modern analytical techniques (e.g. XRF, XRD, XPS, SEM and AFM) is essential to understand target performance under long-term irradiations and to improve the current technology. Moreover, as the design of the target wheels is already similar in the different accelerator facilities and the material is scarce, a common design facilitating experiments with the same targets at different places should be considered.

Nowadays, the expertise on actinide target production as well on their characterization exists in Germany (GSI, Mainz), Russia, Japan (RIKEN) and the United-States. In France, because of the S3 project and physics related to nuclear waste, a laboratory for radioactive targets has been supported by French institutes at Orsay. However, to overcome the current limitations in target technology and for the development of targets that can withstand the high beam currents expected in future accelerators, joint efforts of the different target laboratories will be needed. This research is highly interdisciplinary and thus should be conducted in close collaboration between the target makers, the target users and the accelerator specialists.