EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

UC_x Prototype Target Tests for ActILab-ENSAR

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A. Andrighetto[1], P.Bricault[2], R. Catherall[3], P. Delahaye[7] S. Essabaa[4], H. Franberg-Delahaye[7], A. Gottberg[3,5], I. Günther-Leopold[6] P. Kunz[2], C. Lau[4], B. Roussiere[4], M.G. de Saint-Laurent[7] T. Stora[3], L. Tecchio[1]

[1] *INFN-LNL* [2] *TRIUMF*
[3] *CERN* [4] *IPN-Orsay*
[5] *Université Bordeaux 1* [6] *PSI*
[7] *GANIL*

Spokesperson: T. Stora (thierry.stora@cern.ch)
Co-Spokesperson: A. Gottberg (alexander.gottberg@cern.ch)
Contact person: A. Gottberg (alexander.gottberg@cern.ch)

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Abstract:

Targets based on uranium and thorium refractory compounds have been at the heart of the isotope mass-separation online (ISOL) technique since its first pioneer experiment in 1951. Different developments took place along the years in the various facilities, and today porous uranium carbides with excess graphite phase are used throughout the different operating facilities. However little is known about the influence of their microstructure, crystallography, porosity and chemistry on the isotope release properties. Recently submicron, porous SiC carbide materials with improved exotic sodium and magnesium yields could be used without significant degradation over extended periods at ISOLDE, providing the first direct evidence that such kind of matrices can be used to improve the ISOL beam performance for exotic alkali and earth-alkali beams. Within the framework of ActiLab in FP7-ENSAR: Integrating R&D on ISOL UC targets, several uranium carbide target materials are now under development with the objective to translate recent findings. This will be done by systematic investigations of the impact of phase composition, nano- or submicro-metric grain size, and porosity on their performance and stability. The synthesis are done using different approaches such as carbothermal reduction of oxides and graphite nanometric particles.

Towards this systematic approach a new UC_x target material shall be specifically tested at ISOLDE and is subject of this proposal. Gains are expected for exotic isotopes such as $^{30-32}$Mg, $^{96-98}$Kr, ^{200,229}Rn and ^{200,233}Fr. Most other elements will also likely benefit from this development. The processes of diffusion through the target material and release from the material's surface are particularly complex in the presence of high intensity pulsed proton beam irradiation, which makes it indispensable to perform online measurements. Yields and release curves of representative isotopes will be measured and data will be compared to the results for conventional ISOLDE UC_x targets for the same target and ion source geometry.

Requested shifts: 12 shifts, (split into 2 runs over 2 years)

1 Introduction

Uranium carbide matrices brought to high temperatures (>2000 °C) produce a wide range of radioisotopes through fragmentation, fission and spallation reaction channels. At ISOLDE such targets are brought to interaction with a pulsed proton beam of 1.4 GeV provided by CERN's proton synchrotron booster. Europe's next-generation facilities currently under construction such as HIE-ISOLDE at CERN, SPIRAL 2 at GANIL in France or SPES in Italy will be operated using this principle. The material developed at ISOLDE and now used at RIB facilities around the world is made out of stacked cold-pressed pellets produced by carbothermal reduction of UO_2 and graphite powder. The final stoichiometry is believed to be $UC_2 + 2C$, with coexistence of UC, UC_2 and graphite phases all together with a low uranium density of only $3.5\,\frac{g}{cm^3}$. Although the release properties are expected to be closely related to the material's microscopic structure [1] only little is known about the influence of grain size, microscopic chemical impurities, and microscopic phase coexistence, while only open porosity is a well established parameter for the release of isotopes [2].

2 Scientific Motivation

Recent investigations at ISOLDE have proven that submicron and nanostructured porous materials, such as SiC and Y_2O_3, could significantly improve the release and yields of exotic isotopes such as ^{21}Mg or ^{72}Kr [3]. Material characterizations done or commissioned by CERN reveal coexisting phases of UC_2, UC and C and a large distribution of grain sizes from 3 to 50 μm. Although this material is in use in ISOL-type facilities for several decades only little is known about its microscopic structure both, before but in particular while and after proton irradiation. These effects commonly referred to as ageing cause a major decrease of produced exotic radioisotopes during operation of uranium carbide targets. Figure 1 shows the evolution of ^{30}Na released from different UC_x target units. Besides the large scattering of initial ^{30}Na yields, the figure illustrates the intense fall of production yields of several orders of magnitude within two days. This ageing needs to be addressed in order to be able to provide such exotic beams, intensively requested from uranium carbide targets. It is not known, however, to what extent these ageing effects are caused by degradation of microstructure or by radiogenic chemical evolution. In that way e.g. a quantity of tungsten atoms as an example of a not-released element, is produced from the UC_x matrix during a typical irradiation of $7 \cdot 10^{18}$ protons, which corresponds to an atomic concentration of 10^{-8}, well within typical concentrations, which are known to change macroscopic behavior of materials considerably. In order to pursue on these influences a campaign of micro X-ray adsorption spectroscopy at the Swiss Light Source (SLS) was initiated by the authors for scanning this complex material both before and after proton irradiation on a micrometer scale. The great importance of these investigations was acknowledged by the SLS scientific committee and this activity was granted beamtime already for 2012. Only a comprehensive correlation of the results gained by this spectroscopical technique and from online tests of new UC_x target materials will provide the important missing insights, crucial for the indispensable improvement of uranium car-

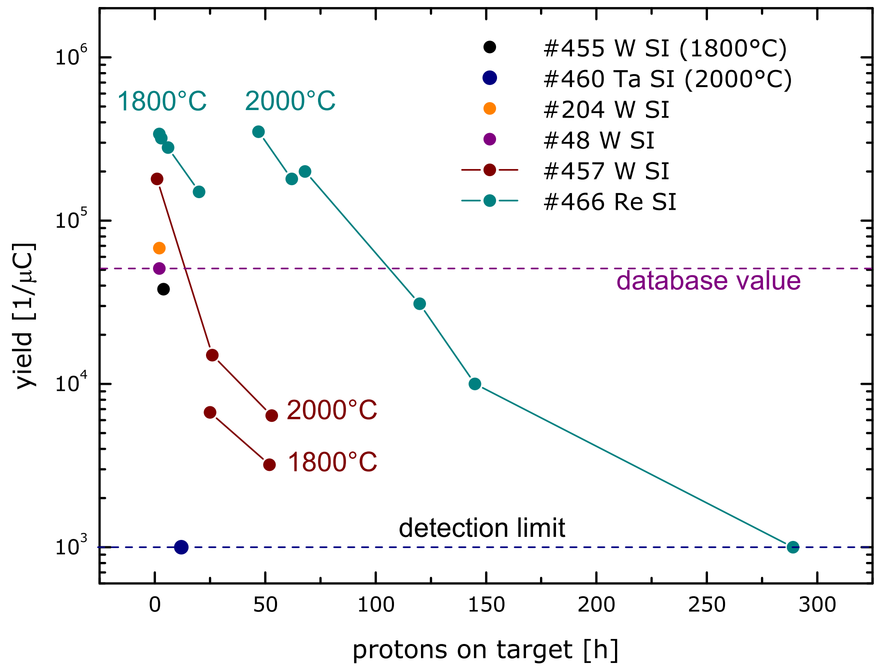

Figure 1: Fluctuations and evolution over time for short-lived radioisotopes extracted from an uranium carbide target, exemplarily shown for ^{30}Na (48ms) [10]

bide targets.
Another recent example (summarized in fig. 2) shows that porous nanostructured calcium oxide has resulted in an increase by an order of magnitude of ^{31}Ar production yields [9]. This enhancement is believed to be easily achievable from uranium carbide targets for exotic noble gases such as Ar, Kr and Rn, but the development of an appropriate uranium carbide material is more difficult due to its radioactivity and toxicity. Changes need to be applied systematically and will be well substantiated by means of various material investigation techniques, such as SEM, BET and XRD at CERN and X-ray absorption spectroscopy, fluorescence spectroscopy and X-ray scattering using a micro focussed synchrotron beam at SLS-PSI. Consequently only online release studies at ISOLDE can reveal the release properties of a new material and are therefore essential towards the development of new exotic radioactive beams.
Towards more defined materials, a uranium monocarbide target was already tested at ISOLDE by an international collaboration in the framework of the 100kW target station Task of F6 EURISOL-DS and later within ActILab in FP7-ENSAR, in October 2010. This material is made of monophasic UC grains from 3 to 9 μm in diameter and a density of 12.3 $\frac{g}{cm^3}$. During these tests the material was found to release alkali and silver isotopes in quantities that did not match the density scaling of x4 between this material and the reference ISOLDE UC_x target, while there were evidences for shorter release times compared to the ones of conventional UC_x material used at ISOLDE [4].

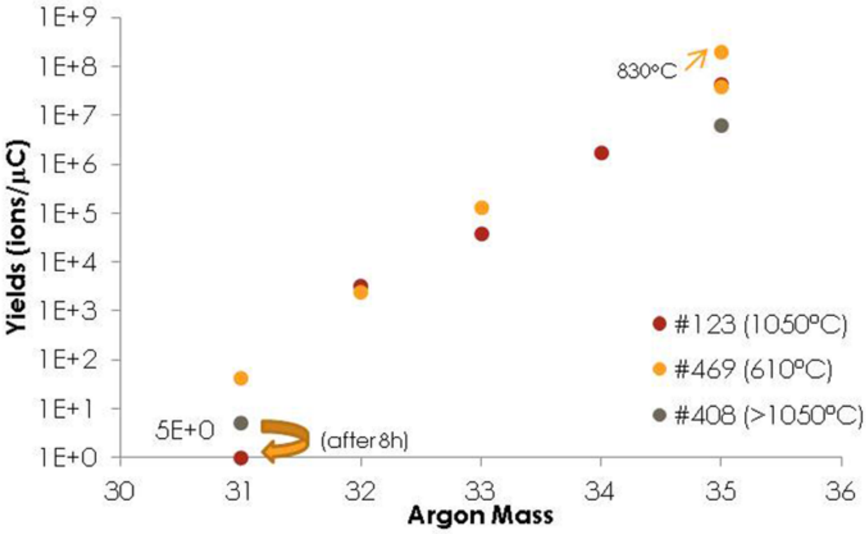

Figure 2: Success in terms of production yields of systematic target material development on the release of Ar isotopes of different half lives. The yellow dots represent the yields measurement from the newly developed nano-grained CaO target material. [9]

To combine the advantages of short diffusion lengths and the high mobility between the material's grains provided by a high degree of open porosity it is now foreseen to perform similar online tests with a UC_x matrix of less than $1\,\mu m$ average grain size synthesized at ISOLDE, matching the optimal characteristics already identified in the innovative SiC matrices. Therefore a methodology similar to the standard ISOLDE preparation is under development, with an UO_2 and graphite preprocessed powder in order to control the particle size, before the cold-pressing takes place. Similarly a new UC_x material has been developed at TRIUMF and comprises a graphite support material for better thermo-mechanical properties [5], this approach shall also be followed up by testing this material at ISOLDE under pulsed beam conditions to assess the impact of time structure on ageing and release properties of this material.
Together with the synthesis of different UC_x materials ongoing in the different institutes part of ActILab, the systematic investigations of their micro-structural and micro-chemical properties, both before and after high intensity pulsed proton beam irradiation, the impact of these tests on the improvement of important exotic isotope production is believed to be verifiable within a few years. It will serve the present program approved at ISOLDE, and the different future facilities such as SPIRAL 2, HIE-ISOLDE and SPES in Europe, and ARIEL in Canada.

3 Tests to Be Performed

Targets and ion sources are tested at ISOLDE in a systematic way with a tape station equipped with a combined beta-gamma detector set-up. Thanks to the pulsed nature of the proton beam available at the PSB, release curves can be obtained for different isotopes with suitable yields and half-lives, figure 3. This can conveniently be fitted by a three exponential function or a more detailed function which takes analytical expressions for diffusion and effusion phenomena into account [6, 11]. Systematic studies

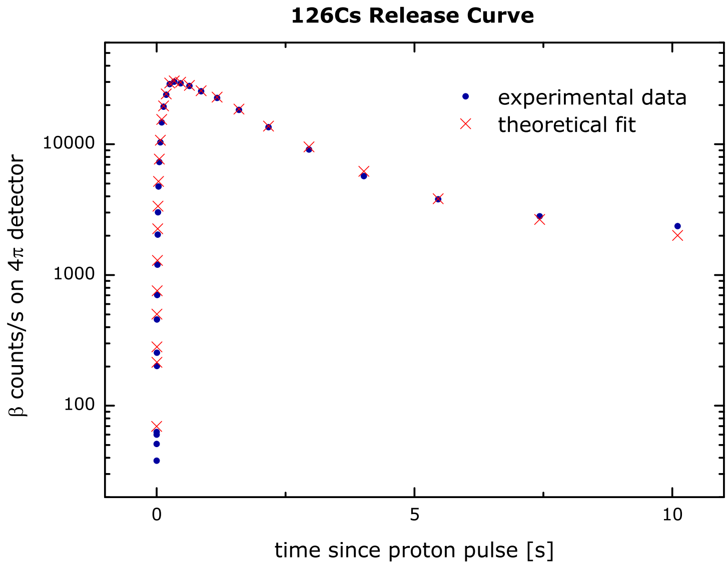

Figure 3: Release function for ^{126}Cs obtained at ISOLDE from a high density UC target brought to 2000 °C, coupled to a tantalum surface ion source. The theoretical points are obtained by the best fit to the experimental points with the three exponential function: $P(t) = \left(1 - e^{-\frac{t}{t_r}}\right) \cdot \left(\alpha e^{-\frac{t}{t_f}} + (1-\alpha)e^{-\frac{t}{t_s}}\right)$, $t_r = 50\,\mathrm{ms}$, $t_f = 2.3\,\mathrm{s}$, $t_s =$ 5.9s, $\alpha = 0.13$

of release properties for a given series of chemical elements is possible thanks to the high production cross-sections provided by fragmentation, spallation and fission reactions of 1.4 GeV protons, which covers most of the periodic table, either on the proton or on the neutron-rich side. It is thus possible to obtain yields and release properties of Li, Na, K, Rb, Cs and Fr alkali isotopes using a surface ion source and of He, Ne, Ar, Kr, Xe and Rn noble gas using a FEBIAD-type ion source coupled to a cold transfer line. The use of the RILIS ion source allows furthermore to selectively ionize diverse other elements of potential interest such as Ag, Sn, Ga or Ni isotopes [8].

Summary of requested shifts:
We envisage to test two different uranium carbide materials in two separate target and ion source units in a standard configuration. In both cases, density, open porosity and stoichiometry will be controlled and tailored. First we will operate an uranium carbide target material, prepared at ISOLDE and as quoted before, coupled to a surface ion source for alkali elements and RILIS for the tests of Ag and Ni isotopes. In a second stage, we intend to test the UC_x+graphite discs composite developed at TRIUMF coupled to a surface ion source to study alkali elements. Tests will done by direct proton irradiation or by neutron induced fissions for suitable elements.

Target and Ion Source	Beam Time
new sub-micro UC_x, ISOLDE design, RILIS, surface ionizer (W), n-converter,	one shift stable beam set-up + one shift to set up RILIS + 5 shifts for radioactive beam tests (direct p-irradiation and n-induced fission)
UC_x on exfoilated graphite TRIUMF design surface ionizer (W), RILIS	one shift stable beam set-up + one shift to set up RILIS + 5 shifts for radioactive beam tests (direct p-irradiation)

4 Conclusion and Summary

It is of primary importance for the various planned or operating worldwide ISOL-type facilities to carry out tests for the quoted newly developed uranium carbide targets in a systematic and standardized approach. The ISOLDE facility offers a unique place to perform such tests thanks to the large available database, the pulsed time structure and to the intense 1.4 GeV proton beam. The performance of these units is expected to be better than the ISOLDE standard units at least for certain isotopes, and therefore appropriate for further use for physics experiments.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT The experimental setup comprises:

Part of the	Availability	Design and manufacturing
"Old" ISOLDE tape station	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Fast ISOLDE tape station	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
SSP-GLM chamber	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT: Hazards named in the document relevant for the fixed SSP-GLM chamber, SSP-GHM chamber installation.

Additional hazards:

Hazards	"Old" tape station	Fast tape station
Thermodynamic and fluidic		
Pressure		
Vacuum	$1 \cdot 10^{-6}$ mbar	$1 \cdot 10^{-6}$ mbar
Temperature	77 K	77 K
Heat transfer		
Thermal properties of materials		
Cryogenic fluid	none	LN2: 1 bar, 100 l
Electrical and electromagnetic		
Electricity	3000 V, 10 mA	3000 V, 10 mA
Static electricity	none	none
Magnetic field	none	none
Batteries	☐	
Capacitors	☐	
Ionizing radiation		

Target material	UC_x	UC_x
Beam particle type	ions	ions
Beam intensity	$\leq 100\,\text{pA}$	$\leq 100\,\text{pA}$
Beam energy	$30 - 60\,\text{keV}$	$30 - 60\,\text{keV}$
Cooling liquids	none	none
Gases	none	none
Calibration sources:	☐	
• Open source	☐	
• Sealed source	☐ [ISO standard]	
• Isotope		
• Activity		
Use of activated material:		
• Description	☐	
• Dose rate on contact and in 10 cm distance	[dose][mSV]	
• Isotope		
• Activity		
Non-ionizing radiation		
Laser	none	none
UV light	none	none
Microwaves (300MHz-30 GHz)	none	none
Radiofrequency (1-300 MHz)	none	none
Chemical		
Toxic	none	none
Harmful	none	none
CMR (carcinogens, mutagens and substances toxic to reproduction)	none	none
Corrosive	none	none
Irritant	none	none
Flammable	none	none
Oxidizing	none	none
Explosiveness	none	none
Asphyxiant	none	none
Dangerous for the environment	none	none
Mechanical		
Physical impact or mechanical energy (moving parts)	none	none

Mechanical properties (Sharp, rough, slippery)	none	none
Vibration	none	none
Vehicles and Means of Transport	none	none
Noise		
Frequency	none	none
Intensity	none	none
Physical		
Confined spaces	none	none
High workplaces	none	none
Access to high workplaces	none	none
Obstructions in passageways	none	none
Manual handling	none	none
Poor ergonomics	none	none

Hazard identification: no hazards involved Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): approx. 2kW per setup