EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Isotope release studies from nanostructured target materials

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Abstract: As part of ongoing advancements in target material development for ISOLtype facilities, nanostructured uranium and lanthanum carbide (UC_x and LaC_x) materials are being investigated to improve radioactive ion beams (RIBs) production. Previous studies on submicron and nanostructured materials have shown notable enhancements in isotope yield and release kinetics, especially for exotic isotopes with short half-lives. With this Letter of Intent we propose to systematically test new nanostructured UC_x and LaC_x materials at ISOLDE, comparing their performance against conventional UC_x and LaC_x targets as as well as a previous nanostructured UC_x material tested in the framework of INTC-P-329 [1]. Gains are expected for exotic isotopes from elements such as In, Mg, Ra, Fr, Sn, Cd, Sb, An, Cu with other elements likely benefiting as well. Isotope yields and release curves will be measured at the tape station using beta and gamma detectors. Ion beam composition analysis and yield measurements will be conducted using the ISOLTRAP Multi-Reflection Time-of-Flight Mass Spectrometer.

Summary of requested shifts: 30 shifts, (split into 3 runs over 1 year)

1 Nanomaterials for ISOL targets

The intensity of a radioactive ion beam at CERN-ISOLDE depends on multiple factors, including the primary beam intensity, target thickness, production cross-section for specific isotopes, as well as the efficiency of isotope extraction, ionization, and purification.

To allow a reasonably fast release of the produced nuclei, the target should be tailored to optimize these parameters by selecting materials that are chemically and thermally stable under operational conditions and facilitate rapid diffusion and effusion rates of the elements of interest [2].

Traditionally bulk and micrometric materials have shown significant performance losses during the release process of more exotic radioisotopes, as challenges related to very short half-lives, low production efficiency and difficulties due to certain elements' refractory nature. The latter issue can be addressed by introducing a chemical reactant to enhance the release of the radioisotopes of interest as molecular beams and provide high-purity beams with less contamination compared to atomic beams [3]. The yields of short-lived isotopes can instead be improved by refining the microstructure of target materials. Previous investigation at ISOLDE have proven that submicron and nanostructured porous materials, such as CaO [4], TiC [5] and SiC [6, 7, 8], could significantly improve the release and yields of exotic isotopes.

For the above reasons, carbides are highly promising for many difficult RIB's, with uranium carbide being extensively used for ion beam production in low and high mass regions. UC_x targets are typically produced by pressing high-purity depleted uranium dioxide mixed with excess graphite, followed by carbothermal reduction under vacuum. A similar process is used for producing lanthanum carbide (LaC_x) targets, with the key difference being that the precursor for lanthanum is its hydroxide $(La(OH)_3)$. These targets exhibit fast release properties and high stability during high-temperature operations.

Over the past decade, significant efforts have been made to further enhance the release properties of exotic isotopes by engineering the microstructure of these materials als [9, 10, 11]. However, due to the complexities involved in producing these materials with stable nanostructure at high temperatures, only one nanostructured UC_x-MWCNT target was tested online at ISOLDE so far. Nevertheless, low-density UC_x (1.4 g/cm³) made from nanometric UO₂ and MWCNT has shown highly promising results, demonstrated superior yields and long-term stability when compared to conventional UC_x (3.5 g/cm³), as highlighted in Figure 1 [1].

The LaCx-MWCNT nanocomposite, previously tested with inconclusive results, will be produced again as part of the commissioning plan for the Class A lab. Its inclusion in the online characterization phase will provide valuable insights into its performance, particularly in terms of release properties and thermal stability. This approach aims to complement the studies conducted with UCx-MWCNT and extend the findings to lanthanum carbide-based systems.

Given these promising results, it would be of significant interest to reintroduce nanostructured UC_x and LaC_x as target materials at ISOLDE. We aim to confirm the yield measurements reported by A. Gottberg [1], and extend these findings with additional measurements in far neutron-deficient and neutron-rich regions.

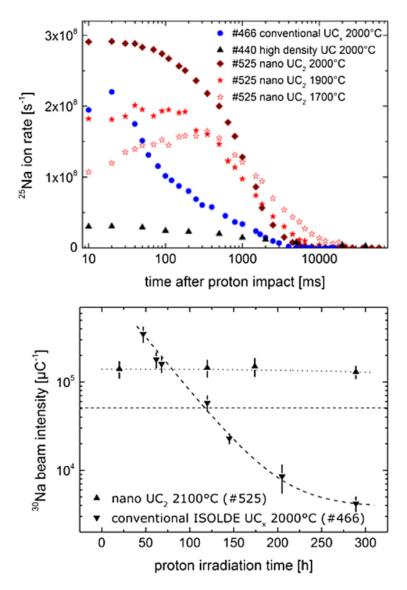


Figure 1: Release time structure of three different UC_x target materials at different operation temperatures (top). Comparison of long-term release stability under high-energy proton irradiation and high temperature (bottom) [1].

2 Regions of interest

We propose conducting three runs with three different targets, structured as follows:

- $\bullet\,$ Target 1: Systematic yield measurements of nanostructured LaC_x target, including RILIS.
- \bullet Target 2: Systematic release and heating measurements of nanostructured UC_x target.
- Target 3: Systematic yield measurements of nanostructured UC_x target.

The following sections provide descriptions of the regions of interest for these targets. For details regarding the shift breakdown, please refer to Section 4.3.

2.1 Lanthanum carbide surface source with RILIS

The neutron-deficient region of the nuclear chart towards the double shell closure at Z, N = 50 is already well-characterized using ISOLTRAP with resonantly laser-ionized beams of In, Sn, Sb, and Cd [12, 13]. The systematic yield and beam composition data available in this region makes it an ideal reference case for a nanometric target material, in addition to the physics importance of the region. Possible contamination such as SrF⁺ can be quantified using the ISOLTRAP MR-ToF MS. In addition to laser-ionized beams, surface-ionized reference isotopes including ^{25,26,30}Na will be used to study release time structures, which can optimally be done during changeover of laser schemes.

2.2 Uranium carbide characterization

A comparative study of the release structures of alkalis will focus on exploring neutrondeficient and neutron-rich chains in Fr and Ra. Reference isotopes, such as 25,26,30 Na and 8,9 Li, for which data from micrometric UC_x targets are available, will be revisited to analyze release time characteristics. In addition to short-lived species of fast-released alkalis, attention will also be directed toward species exhibiting slow release times and low release efficiencies, which remain a subject of interest for future physics investigations at ISOLDE. Among these, actinides are particularly noteworthy.

The isotope 238 U is the nucleus with the highest number of nucleons which is easily available in bulk quantities as a target material, making it virtually the only choice for production of heavy isotopes in the actinide region. Studies of actinide production from standard micrometric UC_x targets [14, 15, 3] have shown that some isotopes of radiogenic Ac, Np, and Pu can be extracted, particularly using molecular extraction. Building on these findings, we aim to check the yields of Ac isotopes.

2.3 Uranium carbide yield measurements

Depending on the outcome of the characterization run, additional yield measurements for nanometric uranium carbide in combination with alternative ion sources may be pursued.

Other species with diffusion-limited release characteristics are also of interest for the running period, including:

- Noble gases (ISOLTRAP) [16]
- Copper (RILIS) [17]
- Bismuth (RILIS) [18]

3 Methods

We will measure isotope production yields and release properties using the fast tapestation located at CA0 [19]. Ion beam composition will be analyzed using ISOLTRAP's MR-ToF mass spectrometer [20] which routinely achieves mass resolving powers on the order of 10^5 and can additionally provide yield data. This combination of decay spectroscopy and mass spectrometry techniques allows us to identify both short and long-lived species over a wide range of isotopes and yields. This technique has been frequently and successfully used for the development of new beams at ISOLDE [3].

References

- Alexander Gottberg. Target materials for exotic ISOL beams. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 376:8–15, Jun 2016.
- [2] Joao Pedro Ramos. Thick solid targets for the production and online release of radioisotopes: The importance of the material characteristics – A review. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 463:201–210, Jul 2020.
- [3] Mia Au. Production of actinide atomic and molecular ion beams at CERN-ISOLDE. Doctoral thesis, Johannes Gutenberg-Universität Mainz, 2023. CERN-THESIS-2023-228.
- [4] J.P. Ramos, A. Gottberg, T.M. Mendonça, C. Seiffert, A.M.R. Senos, H.O.U. Fynbo, O. Tengblad, J.A. Briz, M.V. Lund, G.T. Koldste, M. Carmona-Gallardo, V. Pesudo, and T. Stora. Intense ¹⁰³Ar beams produced with a nanostructured CaO target at ISOLDE. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 320:83–88, Jul 2014.
- [5] J.P. Ramos, A.M.R. Senos, T. Stora, C.M. Fernandes, and P. Bowen. Development of a processing route for carbon allotrope-based TiC porous nanocomposites. *Journal* of the European Ceramic Society, 37:3899–3908, Jul 2017.
- [6] M. Czapski, T. Stora, C. Tardivat, S. Deville, R. Santos Augusto, J. Leloup, F. Bouville, and R. Fernandes Luis. Porous silicon carbide and aluminum oxide with unidirectional open porosity as model target materials for radioisotope beam production. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions* with Materials and Atoms, 317:385–388, Jul 2013.
- [7] P. Hoff, O.C. Jonsson, E. Kugler, and H.L. Ravn. Release of nuclear reaction products from refractory compounds. *Nuclear Instruments and Methods in Physics Research*, 221:313–329, Jul 1984.
- [8] E. Hagebø, P. Hoff, O.C. Jonsson, E. Kugler, J.P. Omtvedt, H.L. Ravn, and K. Steffensen. New production systems at ISOLDE. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 70:165– 174, Aug 1992.
- [9] Julien Guillot, Sandrine Tusseau-Nenez, Brigitte Roussière, Nicole Barré-Boscher, François Brisset, and Sylvain Denis. Development of radioactive beams at ALTO: Part 1. Physicochemical comparison of different types of UCx targets using a multivariate statistical approach. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms, 433(August):60–68, 2018.
- [10] Julien Guillot. Développement de faisceaux radioactifs: Influence de la microstructure d'une cible d'UCx sur les propriétés de relâchement des produits de fission. PhD thesis, L'Universite Paris-Saclay, 2018.

- [11] Julien Guillot, Brigitte Roussière, Sandrine Tusseau-Nenez, Denis Grebenkov, Nicole Barré-Boscher, Elie Borg, and Julien Martin. Development of radioactive beams at ALTO: Part 2. Influence of the UCx target microstructure on the release properties of fission products. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms, 440:1–10, 2019.
- [12] L. Nies, D. Atanasov, M. Athanasakis-Kaklamanakis, M. Au, K. Blaum, J. Dobaczewski, B. S. Hu, J. D. Holt, J. Karthein, I. Kulikov, Yu. A. Litvinov, D. Lunney, V. Manea, T. Miyagi, M. Mougeot, L. Schweikhard, A. Schwenk, K. Sieja, and F. Wienholtz. Isomeric Excitation Energy for ⁹⁹In^m from Mass Spectrometry Reveals Constant Trend Next to Doubly Magic ¹⁰⁰Sn. *Phys. Rev. Lett.*, 131:022502, Jul 2023.
- [13] L. Nies, D. Atanasov, M. Athanasakis-Kaklamanakis, M. Au, C. Bernerd, K. Blaum, K. Chrysalidis, P. Fischer, R. Heinke, C. Klink, D. Lange, D. Lunney, V. Manea, B. A. Marsh, M. Müller, M. Mougeot, S. Naimi, Ch. Schweiger, L. Schweikhard, and F. Wienholtz. Refining the nuclear mass surface with the mass of ¹⁰³Sn, 2024.
- [14] M. Au, M. Athanasakis-Kaklamanakis, L. Nies, J. Ballof, R. Berger, K. Chrysalidis, P. Fischer, R. Heinke, J. Johnson, U. Köster, D. Leimbach, B. Marsh, M. Mougeot, B. Reich, J. Reilly, E. Reis, M. Schlaich, Ch. Schweiger, L. Schweikhard, S. Stegemann, J. Wessolek, F. Wienholtz, S. G. Wilkins, W. Wojtaczka, Ch. E. Düllmann, and S. Rothe. In-source and in-trap formation of molecular ions in the actinide mass range at CERN-ISOLDE. *Nucl. Instrum. Methods B*, 541:375–379, 2023.
- [15] M. Au, M. Athanasakis-Kaklamanakis, L. Nies, R. Heinke, K. Chrysalidis, U. Köster, P. Kunz, B. Marsh, M. Mougeot, L. Schweikhard, S. Stegemann, Y. Vila Gracia, Ch. E. Düllmann, and S. Rothe. Production of neptunium and plutonium nuclides from uranium carbide using 1.4-GeV protons. *Phys. Rev. C*, 107(6):064604, 2023.
- [16] D. Lange, D. Atanasov, M. Benhatchi, K. Blaum, R. B. Cakirli, P. F. Giesel, Y.A. Litvinov, D. Lunney, V. Manea, S. Naimi, L. Nies, C. Schweiger, L. Schweikhard, and F. Wienholtz. Precise mass measurements of light and heavy neutron-rich noble-gas isotopes for nuclear structure studies. Technical report, CERN, Geneva, 2024.
- [17] R.P. de Groote, J.G. Cubiss, C. Schweiger, and K. Chrysalidis. Development of neutron-rich Cu ion beams. Technical report, CERN, Geneva, 2024.
- [18] Andrei Andreyev, Anatoly Barzakh, and Reinhard Heinke. Nuclear and laser spectroscopy study of the neutron-rich ^{212,213,215,216,217,219,220}Bi isotopes with LIST. Technical report, CERN, Geneva, 2023.
- [19] S. Stegemann, D. Atanasov, M. Au, E. Grenier-Boley, M. Butcher, M. Duraffourg, E. Fadakis, T. Feniet, Y. N.Vila Gracia, T. Giles, J. Konki, L. Le, R. Lică, P. Martins, E. Matheson, C. Mihai, R. Martinez Muniz, C. Neacşu, G. Pascovici, K. A. Szczurek, S. Warren, and S. Rothe. The CERN-ISOLDE fast tape station. *Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms*, 541(May):169–172, 2023.

[20] R. N. Wolf, F. Wienholtz, D. Atanasov, D. Beck, K. Blaum, Ch Borgmann, F. Herfurth, M. Kowalska, S. Kreim, Yu A. Litvinov, D. Lunney, V. Manea, D. Neidherr, M. Rosenbusch, L. Schweikhard, J. Stanja, and K. Zuber. ISOLTRAP's multireflection time-of-flight mass separator/spectrometer. *International Journal of Mass Spectrometry*, 349-350(1):123-133, Sep 2013.

4 Details for the Technical Advisory Committee

4.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

- \boxtimes Permanent ISOLDE setup: ISOLDE fast tape station, ISOLTRAP MR-ToF MS, RILIS
 - \boxtimes To be used without any modification
 - \Box To be modified: Short description of required modifications.
- □ Travelling setup (Contact the ISOLDE physics coordinator with details.)
 - \Box Existing setup, used previously at ISOLDE: Specify name and IS-number(s)
 - □ Existing setup, not yet used at ISOLDE: Short description
 - \Box New setup: Short description

4.2 Beam production

For any inquiries related to this matter, reach out to the target team and/or RILIS (please do not wait until the last minute!). For Letters of Intent focusing on element (or isotope) specific beam development, this section can be filled in more loosely.

• Requested beams:

Isotope	Production yield in focal	Minimum required rate	$t_{1/2}$
	point of the separator $(/\mu C)$	at experiment (pps)	
²⁵ Na	TBD*	TBD*	59.1 s
²⁶ Na	TBD [*]	TBD^*	1.07 s
³⁰ Na	TBD*	TBD^*	$45.4 \mathrm{ms}$
⁸ Li	TBD^*	TBD^*	839.9 ms
⁹ Li	TBD*	TBD*	$178 \mathrm{\ ms}$

*To be determined as part of this study.

Other isotopes of interest include:

- Isotopes in the neutron-deficient region of the nuclear chart towards the double shell closure at Z, N = 50
- Neutron-deficient and neutron-rich chains in Fr and Ra
- Ac isotopes
- Cu isotopes beyond $^{78}\mathrm{Cu}$
- Full reference of yield information (e.g. yield database, elog entry, previous experiment number, extrapolation andor justified scaling factors, target number):

- Target ion source combination:
 - Target 1: LaC_x target with Rhenium (Re) surface source
 - Target 2: UC_x target with Tantalum (Ta) surface source
 - Target 3: UC_x target with VD7 or other, depending on the results for Target $_2$
- RILIS? (Yes)
 - Special requirements: No
- Additional features?
 - \Box Neutron converter: (for isotopes 1, 2 but not for isotope 3.)
 - □ Other: (quartz transfer line, gas leak for molecular beams, prototype target, etc.)
- Expected contaminants: SrF^+
- Acceptable level of contaminants: Not applicable
- Can the experiment accept molecular beams? Yes
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? INTC-I-272 and INTC-P-715

4.3 Shift breakdown

Summary of requested shifts:

With protons	Requested shifts
Target 1: Systematic yield measurements LaC_x includ-	8
ing RILIS	
Target 1: reference surface-ionized elements during	3
RILIS element change	
Target 2: Systematic release and heating measurements	8
UC _x	
Target 3: Systematic yield measurements UC_x	8
Target 3: reference surface-ionized elements during	3
RILIS element change	

4.4 Health, Safety and Environmental aspects

4.4.1 Radiation Protection

- If radioactive sources are required: not applicable
 - Purpose?
 - Isotopic composition?
 - Activity?
 - Sealed/unsealed?
- For collections: not applicable
 - Number of samples?
 - Activity/atoms implanted per sample?
 - Post-collection activities? (handling, measurements, shipping, etc.)

4.4.2 Only for traveling setups (not applicable)

- Design and manufacturing
 - \Box Consists of standard equipment supplied by a manufacturer
 - $\hfill\square$ CERN/collaboration responsible for the design and/or manufacturing
- Describe the hazards generated by the experiment:

Domain	Hazards/Hazardous Activities		Description
	Pressure		[pressure] [bar], [volume][l]
Mechanical Safety	Vacuum		
	Machine tools		
	Mechanical energy (moving parts)		
	Hot/Cold surfaces		
Cryogenic Safety	Cryogenic fluid		[fluid] [m3]
	Electrical equipment and installations		[voltage] [V], [current] [A]
Electrical Safety	High Voltage equipment		[voltage] [V]
	CMR (carcinogens, mutagens and toxic		[fluid], [quantity]
	to reproduction)		
	Toxic/Irritant		[fluid], [quantity]
Chemical Safety	Corrosive		[fluid], [quantity]
	Oxidizing		[fluid], [quantity]
	Flammable/Potentially explosive		[fluid], [quantity]
	atmospheres		[iiuid], [quantity]
	Dangerous for the environment		[fluid], [quantity]
Non ionizing	Laser		[laser], [class]
Non-ionizing radiation Safety	UV light		
radiation Salety	Magnetic field		[magnetic field] [T]

	Excessive noise	
Workplace	Working outside normal working hours	
workplace	Working at height (climbing platforms,	
	etc.)	
	Outdoor activities	
	Ignition sources	
Fire Safety	Combustible Materials	
	Hot Work (e.g. welding, grinding)	
Other hazards		