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

Production and Release of Gas and Volatile Elements from Sodium-based Targets

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Abstract

Several large scale facilities being studied for Europe use sodium or a sodium-based alloy either as a target or as a coolant for heavier solid targets subjected to MW proton beams, such as the European Spallation Source (ESS) and beta-beam projects. ESS will be the neutron source in use from the years 2020 in Europe, providing high intensity neutron fluxes over large energy spectra (from 10^{-3} eV to 10^{3} eV) to scientists, to explore materials from 10^{-2} m to 10^{-16} m scale. A sodium-cooled array of tungsten blocks is one of the potential solutions for the target that will convert protons from the 5 MW 2.5 GeV linac into neutrons. Sodium is a tried and tested coolant in fast nuclear reactors with associated technologies and design standards. Its application to a spallation environment however remains to be validated.

The ISOLDE facility is well placed to perform detailed measurements of radioisotopes produced in sodium with a proton beam whose energy of 1.4 GeV is very close to the ESS baseline of 2.5 GeV. We would like to measure the chain of isotopes released from sodium in a controlled experiment. These measurements would lead to an accurate estimation of production cross-sections allowing extrapolation to ESS. The data will contribute to the dimensioning of systems to handle gaseous/volatile elements. It will also enable us to quantify isotopes that will remain in the melt, altering the physico-chemical properties of sodium and establish handling procedures that take into account the residual activity of the sodium and of drained containment structures.

Physics Motivation

Since the approval of the siting of the ESS [1, 2] in May 2009, progress has been made in identifying likely candidates for the target. Options include stationary heavy liquid metal targets and helium- water- or sodium-cooled solid targets. The neutron converter target will receive a pulsed (1 ms, 20 Hz) 5 MW proton beam from the 2.5 GeV Linac. This beam power is similar to the 4 MW proposed for EURISOL [3], the next generation ISOL radioactive ion beam facility in Europe, and goes well beyond the currently achievable ~1 MW at the pulsed spallation source SNS at ORNL.

Collaborating institutes are contributing towards the selection of one solution for the ESS target by carrying out studies that will allow a comparison of the different concepts against an extensive list of criteria. The emphasis is put on engineering feasibility, i.e. the potential for construction, safe operation and maintenance of target systems given materials, heat removal and layout challenges.

A sodium-cooled tungsten target combines the high neutron yield from the densest target material considered for ESS with a coolant that has the highest heat transfer coefficient of all coolants considered. Table 1 shows expected cold neutron fluxes from optimized target/moderator/reflector geometries. As can be seen, tungsten is the best material, even though for other target materials the increased spread of the area from which leakage neutrons exit the target is partially compensated by the effectiveness of the reflector. The final target density to be expected from tungsten is ~90% of its nominal density with some variation from front to back to be defined following detailed coolant flow optimization.

Table 1: *Cold neutron fluxes from an optimized target/moderator/reflector system for different target materials [4].*

Element	Z/A	σ_a (20meV)	I_{res}	Number density	density	$\Phi_{cold@10m}$	$\Phi_{cold@10m}$	$\Phi_{cold@10m}$	Comment
		(barns)	(barns)	(1/barn/cm)	($1/cm^3$)	(n/cm^2/ prot.)	Perf in % vs. Best	Loss in % vs. Best	
W	74/183.8	20	1520	0.063	19.4	6.59E-08	100.00%	0.00%	**Calculated**
Au	79/197.0	110	1940	0.059	19.3	6.05E-08	91.81%	8.19%	**Calculated**
Hg	80/200.6	420	322	0.041	13.5	5.21E-08	79.06%	20.94%	**Calculated**
Tl	81/204.4	28	214	0.035	11.9	5.69E-08	86.34%	13.66%	**Calculated**
Pb	82/207.2	0.5	134	0.033	11.4	5.70E-08	86.49%	13.51%	**Calculated**
PbBi					10.575	5.47E-08	83.00%	17.00%	**Extrapolated**
Bi	83/209.0	0.05	144	0.028	9.75	5.24E-08	79.51%	20.49%	**Calculated**
Pb low	82/207.2	0.5	134	0.033	8	4.56E-08	69.20%	30.80%	**Extrapolated**

At 1.4 GeV, the ISOLDE beam energy is very close to the 2.5 GeV baseline proposed for ESS. In particular, the production rates for most isotopes in sodium are roughly equivalent (see appendix).

Molten sodium salts have recently been proposed for the production of ^{18}Ne in the context of neutrino beta-beam facilities. The proposal outlined here would contribute towards this effort by improving knowledge of production cross-sections in sodium.

Production of hydrogen and helium

Hydrogen and helium will be produced in large quantity during the operation of ESS, and their production and release are of great interest. They will contribute to the total activity of the target station and their extraction from the target, storage in a decay tank and eventual release must be dimensioned. In addition, embrittlement due to hydrogen/helium in structural components will accelerate their degradation.

Production rates for hydrogen and helium from FLUKA [5,6] are presented in table 2 with associated errors expressed as a percentage of production for 1.4 GeV and 2.5 GeV incident proton beams. The production rate for ^{4}He is 0.21 atoms/p, with a statistical error of about 0.3 % to be compared with 0.22 atoms/p at 2.5 GeV.

Table 2: *Production of hydrogen and helium.*

Isotope	A	Z	1.4 GeV atoms/p	Error	2.5 GeV atoms/p	Error
^{1}H	1	1	0.1123	0.1732	0.1207	0.1067
^{2}H	2	1	5.23E-02	0.201	5.98E-02	0.1869
^{3}H	3	1	2.13E-02	0.2153	2.55E-02	0.4143
^{3}He	3	2	2.01E-02	0.3656	2.51E-02	0.3214
^{4}He	4	2	0.2051	0.2643	0.2159	5.28E-02
^{6}He	6	2	1.59E-03	0.9842	1.89E-03	0.9894
^{8}He	8	2	7.30E-05	4.222	1.12E-04	5.583

Production of sodium isotopes

Much larger quantities of sodium isotopes will be produced by spallation with incident protons in the sodium melt compared with the fast reactor spectrum. The most relevant sodium isotopes for activation, maintenance and short-term disposal are ^{22}Na ($T_{1/2}$ = 2.603 yrs) and ^{24}Na ($T_{1/2}$ = 14.96 hrs). They cannot be chemically separated from the sodium melt so production rates must be accurately estimated to determine the activity in any residual sodium throughout the loop.

Table 3: *Production of sodium isotopes.*

Isotope	A	Z	1.4 GeV atoms/p	Error	2.5 GeV atoms/p	Error
^{19}Na	19	11	4.00E-07	61.24	2.00E-07	99
^{20}Na	20	11	1.36E-05	8.575	1.30E-05	13.54
^{21}Na	21	11	8.77E-04	1.915	8.97E-04	0.8002
^{22}Na	22	11	9.54E-03	0.5314	9.58E-03	0.2834
^{23}Na	23	11	5.85E-03	0.6447	6.48E-03	0.5085
^{24}Na	24	11	1.80E-06	47.79	1.80E-06	32.39

Production of noble gas neon

Large quantities of stable (^{20}Ne, ^{21}Ne, ^{22}Ne) and radioactive neon will be produced. The radioactive species (^{17}Ne to ^{26}Ne) either side of stability have half-lives which are ideally suited for measurements at the ISOLDE tape station. Measurements of ^{18}Ne will be of particular interest to the beta-beam community.

Table 4: *Production of neon isotopes.*

Isotope	A	Z	1.4 GeV atoms/p	Error	2.5 GeV atoms/p	Error
17Ne	17	10	2.00E-07	99	0.00E+00	0
18Ne	18	10	2.12E-05	8.621	2.16E-05	7.551
19Ne	19	10	1.79E-04	6.218	1.67E-04	5.988
20Ne	20	10	8.45E-03	0.5255	7.85E-03	0.5336
21Ne	21	10	1.79E-02	0.3527	1.67E-02	0.1887
22Ne	22	10	1.23E-02	0.4035	1.15E-02	0.4331
23Ne	23	10	1.45E-04	1.731	1.60E-04	2.745

Production of oxygen and fluorine

Significant fractions of the neutron-rich oxygen (^{20}O, ^{21}O, ^{22}O) and fluorine (^{20}F, ^{21}F, ^{22}F) isotopes will eventually decay to stable neon. All these will have to be taken into account in estimating the quantities of gas to be handled by the target gas extraction systems. In addition, stable oxygen produced by the beam and by other radioactive elements whose decay chain ends in oxygen will contribute to the chemistry of the residual irradiated sodium system. Oxidation of structural components is likely to be an issue.

Production of other radioisotopes

Other radioisotopes from lithium to nitrogen will also be measured, with the most relevant being ^{7}Be, which might contribute non-negligibly to the total activity within a few days of stopping the beam.

Experimental setup

We propose to run the experiment in two phases, the first with a liquid eutectic of sodium and fluorine such as NaF-ZrF4 and the second with pure sodium. Both targets will use a standard ISOLDE molten metal target container (used for Pb and LBE) equipped with a temperature-controlled condensation helix and connected to a Vadis ion source via a temperature-controlled transfer line. The target container will be filled to ¾ with liquid metal, allowing a free surface for isotopes to diffuse out of the target melt.

Following mass separation, the beams will be detected either with a Faraday cup (e.g. ^{4}He), by on-line β counting and γ spectroscopy with the ISOLDE tape station, or by collection on a foil and off-line γ spectroscopy (e.g. long-lived ^{7}Be).

Accurate knowledge of the ionisation and release efficiencies will be required to estimate cross-sections. The ionisation efficiency measurements for selected elements will be carried out offline by introducing a calibrated leak into the ion source.

The release efficiency depends strongly on temperature and indeed this dependence is of interest to the operation of a spallation target. Specific to ISOLDE is the effect of pulsed proton beam impact, which tends to cause splashing of liquid metals in the target container and a corresponding increase in the release efficiency. At 500°C, the density of pure sodium is 0.83g.cm^{-3}, its volume expansion coefficient is 2.5e-4 K^{-1} and its specific heat is 1.26 J.g^{-1}.K^{-1}. The speed of sound at that temperature is 2300 m.s^{-1} and the isentropic compressibility is 2.24e-10 kg.m^{-1}.s^{-2}. With a peak heat deposition of 25.5 J.g^{-1}, this would correspond to a peak temperature rise and stress in the sodium melt of 20°C and 22 MPa respectively (a factor 40 less than in pure lead [7]). The ratio *s/ρ* was previously defined [7] as a figure of merit to compare proton-generated splashes in different liquid metals. This ratio is 27 for sodium, 61 for tin and 74 for lead indicating that sodium should react much less than either tin or lead targets. Care will be taken in assessing the splashing threshold with experience gained on previous lead targets [8]. In order to account for both temperature dependence and proton beam impact, several runs are required to map the release curve for different target temperature, proton beam intensity and time structure.

Sodium is well known as a coolant in fast reactors. To compare fast reactor and spallation envrionments, a tungsten converter installed below the target would enable measurements of species produced with neutrons only (plus small component of ~100 MeV scattered and secondary protons). Measurements with the neutron converter would precede those with direct beam on target.

Beam Time request

We would like to request 12 shifts online at the GPS for the measurements (6 with the eutectic sodium/fluorine target and 6 with the pure sodium target). A fraction of these measurements can be performed in the GLM/GHM beamlines fully in parallel with another experiment using all remaining proton pulses at the HRS. Only the measurements with the monitoring tape station require the use of the central beamline.

Ionisation efficiency calibration with gas mixtures and stable tracers can be carried out with 2 shifts on the offline separator. The total request is therefore 12 online and 2 offline shifts.

References

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[3] Final Report of the EURISOL Design Study, A design study for a European isotope-separation-on-line radioactive ion beam facility, European Commission Contract No. 515768 RIDS, November 2009.

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Appendix

Isotope production

Production in sodium with a density of 0.83 g.cm^{-3} (density at 500°C) calculated with FLUKA, new evaporation model with heavy fragmentation. The target geometry was taken to be that of an ISOLDE lead target filled to ¾ with sodium. A comparison of two incident proton beam energies on the sodium target is shown in table A.1, 1.4 GeV and 2.5 GeV, with a beam profile of $\sigma_x = \sigma_y = 2.2$ mm. As can be seen, the production rates are very similar at the two proton energies. Data for an incident 1.4 GeV proton beam on a tungsten converter located below the target are also reported.

Table A.1*: Isotope production with beam directly incident on a sodium target at 1.4 GeV and 2.5 GeV and isotope production with beam incident on a tungsten converter located below the ISOLDE target.*

			1.4 GeV		2.5 GeV		1.4 GeV converter	
Isotope	A	Z	/prim	Error	/prim	Error	/prim	Error
1H	1	1	0.1123	0.1732	0.1207	0.1067	1.19E-02	0.5587
2H	2	1	5.23E-02	0.201	5.98E-02	0.1869	2.64E-03	2.436
3H	3	1	2.13E-02	0.2153	2.55E-02	0.4143	7.14E-04	3.849
3He	3	2	2.01E-02	0.3656	2.51E-02	0.3214	2.11E-04	7.005
4He	4	2	0.2051	0.2643	0.2159	5.28E-02	5.79E-03	2.036
6He	6	2	1.59E-03	0.9842	1.89E-03	0.9894	2.13E-05	26.03
8He	8	2	7.30E-05	4.222	1.12E-04	5.583	0.00E+00	0
6Li	6	3	1.20E-02	0.2771	1.29E-02	0.6121	2.02E-04	8.034
7Li	7	3	8.82E-03	0.6229	9.50E-03	0.291	1.58E-04	10.65
8Li	8	3	2.12E-03	1.345	2.39E-03	0.757	3.80E-05	21.18
9Li	9	3	4.04E-04	1.837	4.91E-04	3.709	3.36E-06	66.39
11Li	11	3	7.60E-06	15.34	1.22E-05	15.42	0.00E+00	0
7Be	7	4	4.62E-03	0.5135	5.00E-03	0.8741	6.26E-05	11.21
9Be	9	4	3.65E-03	0.5924	3.65E-03	1.388	8.84E-05	11.65
10Be	10	4	2.03E-03	1.019	2.08E-03	0.712	3.80E-05	18.65
11Be	11	4	1.03E-04	7.187	1.05E-04	1.065	0.00E+00	0
12Be	12	4	2.70E-05	10.08	3.38E-05	9.467	0.00E+00	0
14Be	14	4	0.00E+00	0	2.00E-07	99	0.00E+00	0
8B	8	5	5.26E-04	2.347	6.19E-04	2.029	4.47E-06	51.13
10B	10	5	3.67E-03	0.9537	3.61E-03	0.9872	8.61E-05	8.485
11B	11	5	5.23E-03	0.5839	5.10E-03	0.4651	1.67E-04	11.02
12B	12	5	7.61E-04	1.283	7.28E-04	1.027	2.13E-05	16.71
13B	13	5	2.27E-04	3.502	2.28E-04	2.213	7.83E-06	41.37
14B	14	5	7.00E-06	13.55	8.80E-06	14.1	0	0
15B	15	5	2.00E-07	99	2.00E-07	99	1.12E-06	93.93
9C	9	6	6.32E-05	5.992	7.00E-05	5.44	0.00E+00	0
10C	10	6	2.49E-04	1.564	2.54E-04	2.834	3.36E-06	61.03
11C	11	6	2.37E-03	0.8869	2.46E-03	1.614	4.14E-05	12.63
12C	12	6	1.28E-02	0.501	1.23E-02	0.2221	4.62E-04	5.623
13C	13	6	4.66E-03	0.3639	4.30E-03	0.675	1.83E-04	7.048
14C	14	6	1.35E-03	1.958	1.23E-03	1.293	5.15E-05	14.92
15C	15	6	9.28E-05	2.084	7.72E-05	4.084	2.24E-06	62.09
16C	16	6	8.00E-06	23.39	8.80E-06	23.94	0.00E+00	0
12N	12	7	3.72E-05	4.112	4.08E-05	10.61	1.12E-06	93.93
13N	13	7	4.07E-04	1.513	3.82E-04	1.738	1.57E-05	16.15
14N	14	7	3.21E-03	0.2437	2.97E-03	1.117	1.41E-04	6.661

15N	15	7	5.08E-03	0.5813	4.68E-03	0.9863	2.45E-04	5.505
16N	16	7	8.89E-04	0.864	7.94E-04	2.189	4.14E-05	16.02
17N	17	7	1.05E-04	2.646	8.86E-05	4.876	5.59E-06	37.74
18N	18	7	3.00E-06	27.89	2.20E-06	36.36	0.00E+00	0
13O	13	8	4.80E-06	20.2	7.60E-06	25.17	0.00E+00	0
14O	14	8	4.00E-05	5.303	3.86E-05	5.471	3.36E-06	56.73
15O	15	8	8.73E-04	1.722	8.40E-04	1.695	2.46E-05	19.15
16O	16	8	9.38E-03	0.469	8.81E-03	0.3572	4.09E-04	5.132
17O	17	8	4.54E-03	0.5698	4.19E-03	0.2925	2.51E-04	7.016
18O	18	8	2.39E-03	0.3429	2.12E-03	0.5769	1.67E-04	10.96
19O	19	8	1.19E-04	3.692	9.76E-05	4.566	4.47E-06	37.04
20O	20	8	1.56E-05	7.476	1.68E-05	4.374	0.00E+00	0
21O	21	8	2.00E-07	99	2.00E-07	99	0.00E+00	0
17F	17	9	4.84E-04	2.647	4.69E-04	2.094	2.57E-05	17.24
18F	18	9	4.11E-03	0.5578	3.95E-03	0.8337	1.89E-04	11.01
19F	19	9	1.94E-03	0.7057	1.76E-03	0.7843	2.83E-04	8.263
20F	20	9	1.64E-03	1.177	1.50E-03	1.177	5.82E-04	3.643
21F	21	9	3.89E-04	2.812	3.41E-04	3.056	2.24E-05	22.62
22F	22	9	3.40E-06	40.11	2.00E-06	63.25	1.12E-06	93.93
17Ne	17	10	2.00E-07	99	0.00E+00	0	0.00E+00	0
18Ne	18	10	2.12E-05	8.621	2.16E-05	7.551	0.00E+00	0
19Ne	19	10	1.79E-04	6.218	1.67E-04	5.988	1.01E-05	43.35
20Ne	20	10	8.45E-03	0.5255	7.85E-03	0.5336	4.85E-04	2.705
21Ne	21	10	1.79E-02	0.3527	1.67E-02	0.1887	1.09E-03	2.487
22Ne	22	10	1.23E-02	0.4035	1.15E-02	0.4331	2.08E-03	3.227
23Ne	23	10	1.45E-04	1.731	1.60E-04	2.745	5.16E-04	4.522
19Na	19	11	4.00E-07	61.24	2.00E-07	99	0.00E+00	0
20Na	20	11	1.36E-05	8.575	1.30E-05	13.54	0.00E+00	0
21Na	21	11	8.77E-04	1.915	8.97E-04	0.8002	3.58E-05	12.95
22Na	22	11	9.54E-03	0.5314	9.58E-03	0.2834	8.09E-04	2.714
23Na	23	11	5.85E-03	0.6447	6.48E-03	0.5085	2.64E-02	0.4117
24Na	24	11	1.80E-06	47.79	1.80E-06	32.39	1.01E-05	34.91
21Mg	21	12	4.00E-07	61.24	0.00E+00	0	0.00E+00	0
22Mg	22	12	1.36E-05	14.07	1.10E-05	7.606	0.00E+00	0
23Mg	23	12	1.27E-04	3.486	1.05E-04	3.113	1.90E-05	16.32
24Mg	24	12	0.00E+00	0	4.00E-07	61.24	0.00E+00	0
26Al	26	13	2.00E-07	99	0	0	0.00E+00	0