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Comparison Of Expected Yields For Light Radioactive Beams At SPIRAL-1 And 2

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Abstract. Light-particles stable high-intensity beams (p, d, ^{3,4}He...) will be available from the Linac driver accelerator of SPIRAL-2. Yields of neutron-deficient isotopes as well as of neutron-rich isotopes (by means of reactions with secondary neutrons) are compared with those presently available at the SPIRAL-1 facility. We explore, for light beams (Z < 16) asked in the 'white book' of SPIRAL-2¹, the production methods taking into account the in-target yield but also the feasibility of making such beams by the ISOL method (considering reaction, target, thermal and release properties). We discuss some of the tests needed and planned. A comparison with the present and potentially attainable yields at SPIRAL-1 is presented.

Keywords: Reaccelerated radioactive beams, Sources of radioactive nuclei, Nuclear reactions

PACS: 29.38.Gj, 29.25. Rm, 24

INTRODUCTION

A major goal of SPIRAL-2^{2,3} is to produce neutron-rich heavy radioactive beams using natural-uranium fission. Neutrons are generated by deuteron stopping in a thick target, the so-called converter. The new superconducting LINAC accelerator, driver of SPIRAL-2, shall accelerate other light and heavy ions with high intensities. Fusion-evaporation residues, transfer or deep-inelastic products, using n, p, d, ^{3,4}He and heavy-ion beams can thus be produced using a variety of targets. Two working groups have been formed for the "SPIRAL-2 Preparatory phase"⁴ in the context of the 7th Framework program (FP7): one (task 7.1) is composed of GANIL (France), Soreq Nuclear Research Center and Weizmann Institute (Israel) physicists. Its role is to study the production options for the intense light ion beams listed above. Another working group (task 7.2) is composed of CENBG-Bordeaux and IFIN-HH-Bucarest with the aim to study the production of radioactive beams induced by heavy ions.

The subject of this contribution is the comparison of yields expected for light radioactive beams at SPIRAL-1 and after an upgrade permitting new fragmentation reactions and those at SPIRAL-2. The goal of the study is to find out what is the most promising alternative in order to concentrate development efforts on it. We review

nuclear reactions induced by p, d, ^3He , ^4He (with energies up to 33, 40, 72.5 and 80 MeV, respectively, corresponding to the Linac maximum energy ⁵) and those induced by secondary neutrons generated by a 40 MeV - 5 mA deuteron beam on a graphite thick target.

STATUS OF RADIOACTIVE BEAMS AT SPIRAL-1

Available Radioactive Beams at SPIRAL-1

The present SPIRAL-1 uses heavy-ion projectile fragmentation, a graphite target and a standard N^+ -ECR ion source, equipped with a cold transfer pipe between target and ion source. The radioactive beams available after acceleration are $^6,^8\text{He}$, ^{13}N , $^{14,15,19-22}\text{O}$, $^{18,20-21}\text{F}$, $^{17-19,23-27}\text{Ne}$, $^{31-35,42-46}\text{Ar}$ and $^{72-77,79,81}\text{Kr}$ ⁶. The cold transfer pipe traps non-volatile elements and therefore allows for a high degree of beam purification, transmitting only noble gases and volatile molecules stable at high temperature to the ion source. However, with such device, non-volatile beams are not possible and effusion time reduces the efficiency of volatile elements with short radioactive period.

Tests with radioactive 1+ ions

Extensive R&D on the extraction of non-gaseous elements as 1^+ ion has been carried out at GANIL using the on-line test bench SIRA separator. An ECR-1+ ion source without cold transfer pipe ⁷ and a Surface Ionization Source ⁸ were tested. The nuclei $^{32,33,34}\text{Cl}$, $^{30,31}\text{S}$, $^{29,30}\text{P}$, $^{28,29}\text{Al}$ and $^{21,22,23,27}\text{Mg}$ have been produced using ^{36}Ar projectile fragmentation ⁷ and ionized with the ECR. The total efficiencies (diffusion-effusion, 1+ ionization, extraction at 20 kV and transport) were between 3×10^{-2} and 1×10^{-4} , depending on the isotopes. Hydrides (HCl, HS, HP, due to residual vacuum) were also observed. Keeping stable intensity during a long running period has still to be demonstrated. The yields could be improved using dedicated incident beams (^{32}S , ^{28}Si or ^{24}Mg) leading to greater cross-sections. During the tests, the ion source was sensitive to the C target out-gassing and difficult to tune.

The nuclei ^{47}K , $^{25,26,27,28}\text{Na}$, $^{28,29,30,31}\text{Al}$ and $^8,^9\text{Li}$ have been produced using ^{48}Ca projectile fragmentation ⁸ and 1+ ionized by surface ionization in a carbon pipe. The total extraction efficiencies depend of the isotopes. They are from $4.3 \cdot 10^{-1}$ for ^{25}Na to $1.6 \cdot 10^{-2}$ for ^{28}Na . These yields could be improved using a more suitable primary beam (^{26}Mg , ^{30}Si or ^{36}S).

In general the target adaptation with a 1+ ion source (ECR with a bigger plasma volume and without transfer pipe, surface ionization source, or hot Febiad with hot transfer tube) taking into account the available space in the storage shielding remains to be done ⁹.

Possible RIB's at SPIRAL-1

The accelerated and 1+ beams mentioned above are first steps towards new beams at SPIRAL-1. In particular the release out of C target has been demonstrated for the elements He, Li, N, O, F, Ne, Na, Mg, Al, P, S and Cl. In the case of non-volatile elements an appropriate ion source has to be used. Release has to be demonstrated for Be, C, B and Si: Beryllium beams are produced with a UC2/graphite target at ISOLDE¹⁰ and a Ta target at TRIUMF¹¹. Radioactive carbon ions cannot be released without exchanging the C-target material. An authorization from safety authorities is needed to change the nature of the target at SPIRAL-1. Finally, the elements boron and silicon have never been, to our knowledge, extracted as radioactive isotopes by the ISOL method.

Post-acceleration with the cyclotron CIME requires a high charge state. The 1+ ion source will be modified in the context of a new 1+/N+ breeder at SPIRAL-1⁹.

NEUTRON-DEFICIENT BEAMS AT SPIRAL-1 AND SPIRAL-2

The in-target yields (particles per second) expected at SPIRAL-2 for ^{14,15}O, ¹⁸F, ^{18,19}Ne, ^{21,22}Na, ^{22,23}Mg, ²⁶Si, ²⁹P, ^{30,31}S, ³³Cl are reported in fig. 1. The primary beams are p, d, or ^{3,4}He with assumed current of 100 μAp. The targets are: C, MgO, Al₂O₃ oxides, and the SiC carbide, all known for their refractory character. They are already used at ISOL installations, although at much lower deposited power. Yields shown are calculated using experimental cross-sections taken from literature (open circles) when existing or from the simulation database EAF07¹² (squares), or the code PACE4¹³ (triangles). When using cross-sections the following equation (eq.1) is used

$$Yield = I \cdot \int \frac{dx}{dE}(E) \cdot \sigma(E) \cdot dE \quad (1)$$

Only reactions leading to the best yields are reported. Some 10¹⁰ to 10¹¹ pps are expected. Exceptions are ²²Mg and ³³Cl for which no reactions using solid targets in the available energy range of primary beams were found. Yields in the SPIRAL-1 graphite target are also calculated (filled circles) using the EPAX parameterization version 2¹⁴, at 1.5 kW. For 100 μAp, a factor 10 in favor of SPIRAL-2 is observed, but it does not take into account the problem of beam power dissipation in the production target. Yet, efficiency of products release has not been considered.

Production of ¹⁴O at SPIRAL2

The oxygen isotopes ¹⁴O and ¹⁵O have been identified as the neutron-deficient light beams to be investigated with highest priority for SPIRAL-2¹. We discuss production of ¹⁴O in some detail.

Cross Sections

The reaction which has the largest cross-section to produce ^{14}O is $^{14}\text{N}(p,n)^{14}\text{O}$ ¹⁵. However, it implies release of oxygen from gaseous N target or from a solid BN target. Oxygen release out of BN target has not been observed at Louvain-La-Neuve¹⁶. The $^{12}\text{C}(^3\text{He},n)^{14}\text{O}$ and $^{12}\text{C}(^4\text{He},n)^{15}\text{O}$ reactions have therefore been investigated for a first study for SPIRAL-2. A solid graphite target allows $^{14,15}\text{O}$ extraction via the CO molecule. Unfortunately, yields deduced from experimental cross-sections¹⁷ and those directly measured for a thick-target¹⁸ differ by a factor 7. At SPIRAL-1 the reaction used to produce this isotope is ^{16}O projectile fragmentation on a C target. An in-target yield gain, 10 to 70 times depending on the actual cross-section, could be achieved at SPIRAL-2 with respect to SPIRAL-1. This estimate is calculated using realistic conditions implying the present 1.5 kW target for SPIRAL-1 and a 5 kW-target (30 MeV, 0.16 mA, ^3He) for SPIRAL-2. Because of the factor 7 of uncertainty for SPIRAL-2, the in-target $^{14,15}\text{O}$ cross-section will be measured at the Nuclear Physics Institute Rez (Czech Republic)¹⁹.

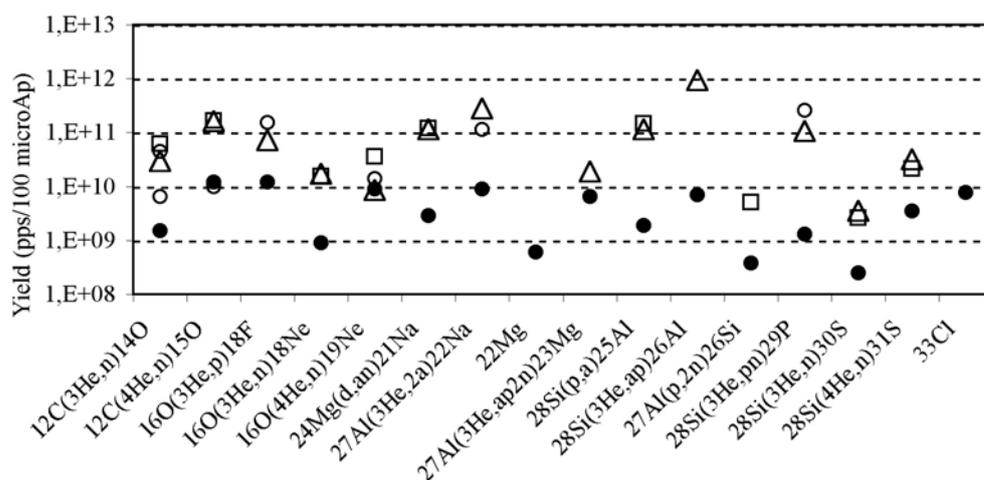


FIGURE 1 Expected yields (pps) for SPIRAL-2 using 100 microAp p, d, or $^3,^4\text{He}$ calculated using cross-sections from literature (open circles) from PACE4 code (triangles) and simulation database EAF07 (squares) and yields in the SPIRAL-1 C target (filled circles).

Thermal Issues

In order to dissipate high beam power, the primary beam has to be defocused on the target, implying the use of large radius and thin target material. Thermal tests of C-foils have been performed at AIFIRA at CENBG, Bordeaux, using their on-line pyrometers with a 1 MeV proton beam. Figure 2 shows the foil temperature dependence on beam intensity. Diamonds, circles and squares correspond to 3 different pyrometers suitable in different temperature range. The influence of target

thickness for the same flexible foil material is shown in fig. 2a. Two graphite types of same thickness do not exhibit large temperature differences (fig. 2b).

Thermal simulations are being performed at GANIL in order to determine how much primary 35 MeV ^3He power can be sustained by the C-target. The ^3He range in C is 0.74 mm (for $\rho= 1.8\text{g}/\text{cm}^3$). In a first approach, we have limited the target box size to the space available for the UCx container of SPIRAL-2. A schematic drawing of the C-target (fig 3) was simulated using the code Comsol Multiphysics²⁰ in 2D with axial symmetry axis. The stainless-steel box was cooled by water assuming a thermal conductivity coefficient $k=4.45 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. Temperature was calculated as a function of the primary beam intensity (fig 4a). The graphite target for a flat beam of 4 cm radius can accept an intensity of 200 μA . Influence of beam shape has also been explored (fig 4b). It shows the importance of defocusing and flatness of the ^3He beam. The calculation predicts a temperature increase of 300 K at $I= 100 \mu\text{A}$ (800 K at 500 μA) if a Gaussian beam profile with 90% of intensity within 4 cm target radius is used instead of a flat one (fig 4b). These preliminary simulations will be pursued using a more realistic target box drawing.

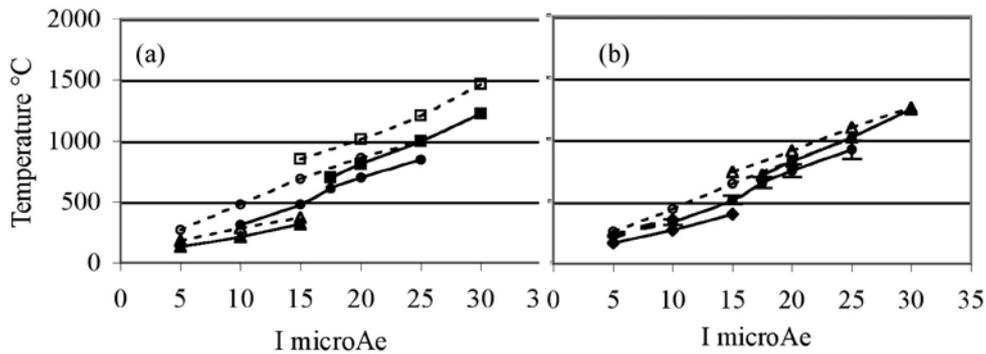


FIGURE 2 : Temperature in graphite foils versus 1 MeV proton intensity: a) for flexible C foils of thickness 125 (filled symbol) and 75 μm (open symbol), b) for flexible (filled symbol) and rigid (open symbol) C foils both of 125 μm thick.

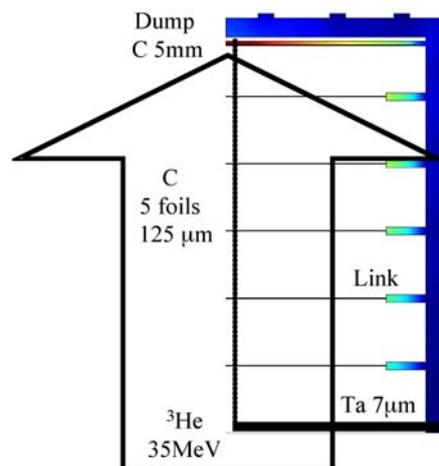


FIGURE 3 : Schematic drawing of the simulated C-target (only half of target is shown).

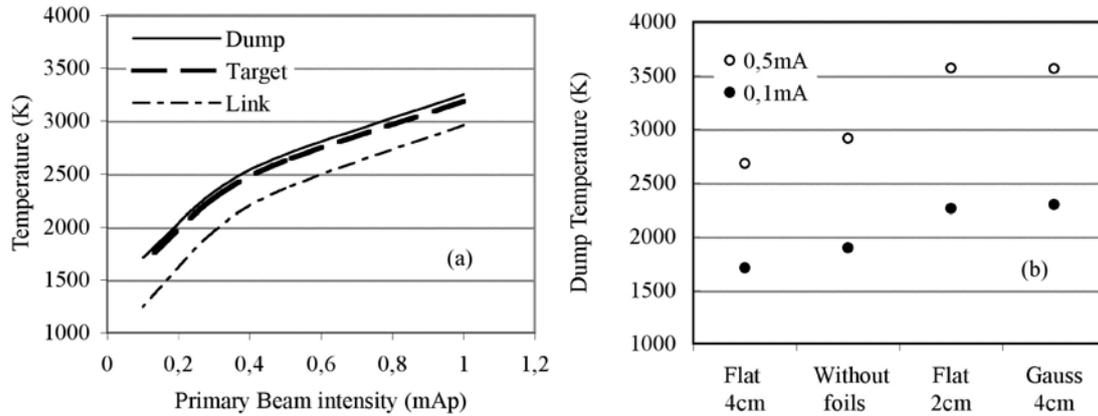


FIGURE 4 : a) Temperature as function of the primary beam intensity for a flat 35 MeV-³He beam of 4 cm radius, b) Dump temperature for different beam profiles.

NEUTRON-RICH BEAMS AT SPIRAL-1 AND SPIRAL-2

The neutron-rich nuclei ⁶He and ⁸Li were chosen to be investigated with highest priority for SPIRAL-2 and SARAF. We discuss also our attempts to find a production scheme for ¹⁵C and nitrogen isotopes. These isotopes shall be produced by neutron-induced reactions. Neutrons will be produced by deuteron break-up, stripping or (d,xn) evaporation in a first target called neutron converter. The targets we envisage could be placed instead of the UCx target behind the converter. The 1500°C-oven developed for SPIRAL-2³ would be used to favor the release of reaction products. In the presently adopted scheme, based on fully stopping 40 MeV deuterons in carbon, the spectrum of neutrons impinging on such a target has an energy-integrated conversion factor close to 1% of the primary beam intensity and has the following distribution: 50% of them are below 10 MeV, 40% between 10 and 20 MeV, and 10% between 20 and 40 MeV. More details can be found in Ref.²¹.

⁶He production

The reaction selected to produce ⁶He (0.8 s) is ⁹Be(n,α). T. Hirsh et al.²² from the Weizmann institute performed series of simulations using the Monte-Carlo MCNP4b code. An in-target production of 2.10¹³ pps is expected at SARAF using a 2 mA, 40 MeV deuteron beam in truly optimal conditions (Li converter for neutron production, natural density BeO, without spacing in the volume target). The strong ⁹Be(n,2n)⁷Be reaction doubles the number of useful neutrons²².

At the beginning stage at least, the SPIRAL-2 converter will be a rotating C wheel. This lowers the neutron flux by a factor of 2 with respect to the Li-converter²³. The BeO material will have 70% of natural density due to the porous μm-structure necessary for He release. Moreover, the available space shall be filled to about 2/3 due to the crumbly nature of the pellets. Thus the apparent density will rather be between 33% and 50% of the natural one. Lower density reduces the neutron flux enhancement via ⁹Be(n,2n)⁷Be down to 1.2 (if ρ/3) to 1.4 (if ρ/2). An in-target ⁶He rate of 4.10¹¹ pps can be obtained from the convolution of the known experimental cross-section²⁴ with

the measured neutron distribution²¹, and assuming an effective density of $\rho/3$ and 1 mA deuteron current at the beginning of operation of SPIRAL-2. However this calculation has not taken into account the channels $(n,t+p)$, $(n,^3\text{He}+n)$, $(n,d+d)$, $(n,2p2n)$ which are opened above 20 MeV but whose cross-sections are not known experimentally. This rate, while much lower than the yield calculated for SARAF, is still 50 times the one at SPIRAL-1 (8×10^9 pps at 2.5 kW). Let us then further remark the extracted rate at SPIRAL1 of 2×10^8 pps without acceleration and 3×10^7 pps to 5×10^6 pps after acceleration depending on final ^6He energy.

Figure 5a shows the phase diagram of BeO calculated at GANIL with F*A*C*T code²⁵ when the target is alone or when it is in contact with C, Ta, Mo, W or Re. Only W or Re do not reduce in the pressure-temperature range where BeO is in solid phase. Moreover at low pressure, the allowed temperature decreases. Off-line thermal tests of material at CERN-ISOLDE have shown that Re is the best choice (fig 5b). A collaboration²⁶ (ISOLDE, GANIL, SOREQ) has recently performed an on-line experiment using neutrons on a 100 g BeO target to produce ^6He . A rhenium foil separated the BeO from the tantalum container. The ^6He production was comparable with the one at low energy at SPIRAL-1. The analysis of release efficiency is in progress.

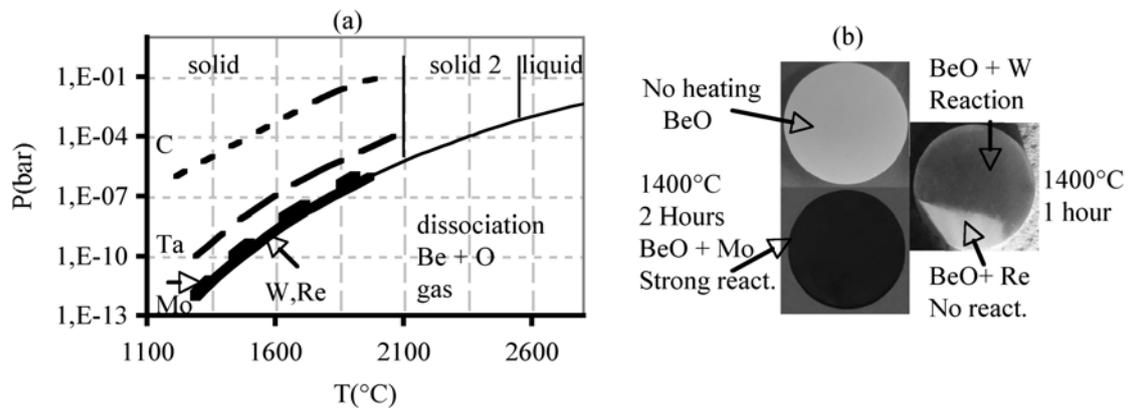


FIGURE 5 : (a) Phase diagram of BeO calculated with F*A*C*T code when alone (thin line) or in contact with C, Ta, Mo, W or Re. W or Re do not reduce the pressure-temperature range of BeO in solid phase. (b) Off-line thermal test at Isolde : BeO alone at room teperature, in contact with Mo,W and Re inserted in Ta boat at 1400°C.

^8Li production

As for ^6He , extensive simulations^{15,22} have been performed by the SOREQ group concerning ^8Li production by $^{10}\text{B}(n,\alpha)^8\text{Li}$. In-target rate of $8 \cdot 10^{12}$ pps is expected at SARAF assuming 2 mA deuterons, Li converter, and natural B target density. The yield at SPIRAL-2 is expected to be $1.4 \cdot 10^{11}$ pps using 1 mA deuterons, C converter, and a BN target, which corresponds to 20 times the in-target SPIRAL-1 yields ($7 \cdot 10^9$ pps at 1.5 kW). However, Li is the most difficult alkali to be ionized. The $^8\text{Li}^+$ efficiency as measured on the SIRA test bench separator is only $5 \cdot 10^{-5}$ pps. Off-line thermal tests are in progress at SOREQ with a recently acquired BN material with μm -

structure. Material choice and diffusion measurement will be especially important since the ^8Li half-life is 0.8 s only. We note that stable Li has been released from BN with a short component of a few seconds²⁷. B_4C material will be also investigated.

^{15}C and $^{16,17,18}\text{N}$ production

In general, neutron-rich nuclei could be produced by neutrons using various targets placed behind the converter. For instance, ^{15}C , $^{16,17,18}\text{N}$ rates have been calculated using MCNP code at GANIL. The target is Al_2O_3 ($\rho = \rho_{\text{natural}}/3$). Figure 6 presents the expected yields calculated using cross-sections from MCNP, Pace4, or the simulation database EAF05. The available neutron energies are well overlapping with cross-sections for (n, α) channels, but $^{18}\text{O}(n,\alpha)^{15}\text{C}$ is not suitable as a big target due to the small natural abundance of ^{18}O (<0.2%). The neutron spectrum is too low in energy for efficient use of $^{16}\text{O}(n,2p)^{15}\text{C}$. Alternatively, ^3He up to 72.5 MeV should be available at the Linac. Higher-energy neutrons could be therefore produced by $\text{C}(^3\text{He},xn)^{28}$. Unfortunately, the neutron yields (in the 20-40 MeV range needed for (n,2p)) are of the same order as with 40 MeV deuterons on carbon. Charged-particle reactions leading to ^{15}C are $\text{B}^{15}\text{N}(d,2p)$ or $\text{B}^{14}\text{N}(t,2p)$. The former, due to low ^{15}N natural abundance, leads to $7 \cdot 10^7$ pps in the target. The cross-section of the second one is not known and it is in any case delicate to use a high-intensity tritium beam.

At SPIRAL-1 $^{15,16}\text{C}$ are easily produced in the target using ^{18}O projectile fragmentation. However, they cannot be released without evaporating the graphite target. This calls for another target than graphite. A metal oxide target allows release of $^{15,16}\text{CO}$ molecules. A first test at SIRA-GANIL²⁹ has demonstrated the $^{11,15,16}\text{C}$ extraction using only 100 nAe, 25 MeV/A ^{22}Ne beam and Al_2O_3 , HfO_2 , ZrO_2 targets. However, metal oxides in normal conditions of temperature and pressure are insulators. The stopped $^{18}\text{O}^{\text{N}+}$ ions shall charge the target, possibly leading to discharge and consequent physical damage of the target. Noting that $^{15,16}\text{C}$ fragments have a higher range than the oxygen ions beam, a target in two parts could be a solution. The first part in graphite for radioactive production should be thick enough to stop the ^{18}O beam, but not the lighter-Z products. The second part in metal oxide acts as a catcher for lighter products and enables $^{15,16}\text{C}^{\text{stable}}\text{O}$ molecule formation. This solution is rather complicated (separation of C and oxide parts by a window, need for an oven to heat the oxide, incomplete separation per range, etc).

Fragmentation of ^{18}O , ^{22}Ne or ^{26}Mg projectiles could be used for $^{16,17,18,19}\text{N}$ production. The in-target yields calculated with the EPAX parameterization is from 10^{10} pps for ^{16}N down to $3 \cdot 10^7$ pps for ^{19}N . An accelerated ^{13}N (9.9 m) beam is already available at SPIRAL-1 using a standard NANOGAN N+ ECR ion source with a cold transfer pipe. The measured efficiencies are: $7 \cdot 10^{-3}$ for $^{13}\text{N}^{1+}$, $5 \cdot 10^{-4}$ for $^{13}\text{N}^{2+}$ and $\text{C}^{13}\text{N}^{1+}$. CN molecule formation helps probably the N release from graphite μm -structure.

The cold transfer pipe between target and ECR is probably a handicap for best efficiency for such short-lived isotopes ^{16}N (7 s), ^{17}N (4.2 s), ^{18}N (0.6 s) and ^{19}N (0.3 s) and molecule stability. A 1+ ion source without cold pipe could be a better solution¹¹.

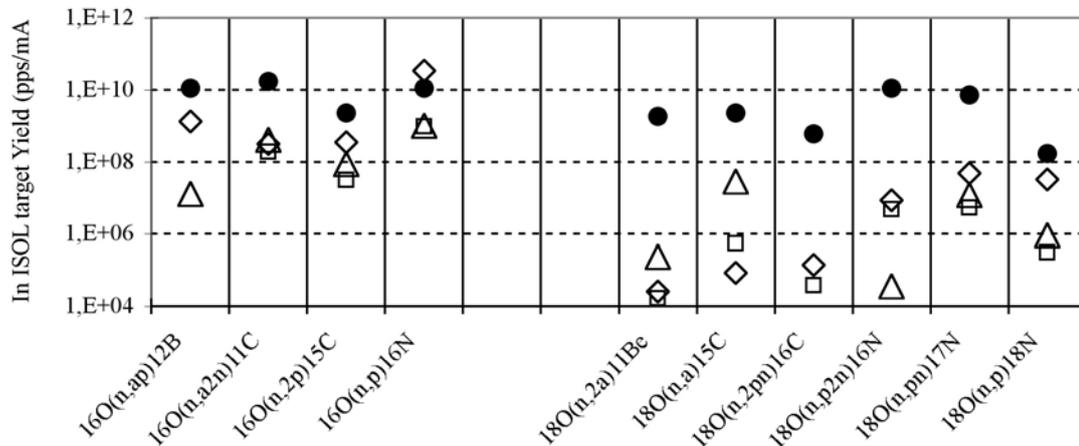


FIGURE 6 Expected yields (pps/1 mAp of deuteron beam on carbon converter) calculated using MCNP (diamonds) or cross-sections from PACE4 code (triangles) and simulation database EAF05 (squares). Yields inside SPIRAL-1 C target are also calculated (filled circle) using the code EPAX¹⁴ and assuming 1.5 kW.

CONCLUSION

The in-target yields at SPIRAL-2 for light-charged particles p, d, and $^3\text{-}^4\text{He}$, and neutrons impinging on low-mass element targets have been estimated and compared with existing or potentially reachable values at SPIRAL-1.

For neutron-deficient nuclei a gain of yield in the targets of about a factor 10 is expected assuming that the SPIRAL-2 target could withstand 100 μA of light-charged particles. Thermal behavior and reaction product release remain challenges to be solved. The targets MgO , Al_2O_3 , SiC are already used at ISOL installations but not at such high power. The first thermal studies and simulations performed at CENBG and GANIL for $\text{C}(^3\text{He},n)^{14}\text{O}$ shows that 200 μA of 1 MeV protons can be accepted by a C-foil target and shows the importance of defocusing and flatness of the primary beam profile.

For neutron-rich nuclei, a gain of about a factor 100 is expected for ^6He , ^8Li when a 5 mA deuteron beam will be available. SPIRAL-1 should be competitive for the nuclei $^{15,16}\text{C}$, $^{16,17,18}\text{N}$ and ^{25}Ne . At SPIRAL2, an in-target gain of factor 10 is expected for ^{23}Ne , $^{25,26}\text{Na}$ using 1 mA deuteron apart from any consideration of losses due to release. Experimental cross-sections for energetic neutron reactions, especially above 20 MeV, are scarce and often in disagreement with each other. It is further to be noted that neutron targets are very thick, making prompt release an even more harder-to-solve challenge than for n-deficient nuclei.

Finally, high charge states are required for post-acceleration in the CIME cyclotron; the $1+/N+$ breeding efficiency for such light nuclei is another challenge.

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