RNB production using a powerful proton accelerator

D. Ridikas[1*], W. Mittig[2], N. Alamanos[1]

1) CEA Saclay, F-91191 Gif-sur-Yvette, France
2) GANIL, BP 5027, F-14076 Caen, France

Abstract

The aim of the considerations below is to estimate the orders of magnitude for the production of intensive RNBs by the use of the powerful proton accelerator **if available**. We try to take into account present technical limitations of ISOL-techniques by limiting our discussion to the **production of fission products**. These possibilities are compared to existing or projected RNB facilities.

1 Introduction

A significant increase of the intensities of RNBs will certainly open access to a new domain of physics. Future extension of the present facilities or the construction of new facilities would allow to reach RNBs with the intensities of 10^{12}pps, and thus to study the cross sections down to the μb or even nb level.

Here we assume that a powerful proton accelerator (∼1GeV; a few MW) is available, what is about 3 orders of magnitudes more than currently used in any ISOL and fragmentation of the beam facilities, all below 10kW power limit. On the other hand, proton beams of ∼160kW are already available at RAL (UK) and more than 1MW at PSI (Switzerland). The research to develop high-intensity beams of several MW is in progress, mainly stimulated by the projects such as Accelerator Transmutation of Waste (ATW), Spallation Neutron Sources (SNS), Accelerator Production of Tritium (APT), etc.

Unfortunately, an increase of primary beam intensity does not necessarily increase the intensity of the secondary RNBs as was shown in [1]. The authors argue that the maximum RNBs will be obtained actually for a limited incident beam power. This is an important warning that there may be quite severe limitations with respect to the admissible power on the target. For the RNB facilities based on the charged particle induced reactions, this limitation is of the order of ∼20-30kW in the RNB production target [2]. An alternative way of producing the RNBs in the mass region of 75<A<160 can be achieved utilizing a target-converter (neutron source) [3]. The emitted neutrons then interact with a fissionable target. Contrary to the charged particles, the neutrons will heat the target indirectly and mainly by the "useful" fission reactions.

*ridikas@cea.fr

In this context we will discuss mainly two possibilities: a) the neutron induced fission, where the neutrons are produced in a converter from a primary proton beam; b) the proton induced fission/spallation, where the primary beam directly hits a heavy target. The major goal is to reach at least 10^{14} fissions/s in order to be compatible with reactor-driven or other future RNB facilities. Another equally important possibility – a spallation of intermediate mass targets that may provide very broad range of isotopic production – will be considered elsewhere. We note in this context that the isotopic distributions of low energy fission and high energy spallation are very different; a high energy spallation provides a very broad production range, whereas a low energy fission gives the highest yields in the region covered by this process.

We employ a coupled LAHET+ MCNP+ CINDER code system [4] for all numerical calculations presented in this work.

2 The present and projected RNB facilities

The production targets for the future ISOL facility should be able to work at high beam power, and at the same time, be coupled efficiently to ion sources for the production of the secondary beams of short-lived isotopes. Beam power of 100kW or more involves extrapolation beyond current experience at any ISOL-type facility. Indeed, the highest charged particle beam power dissipated so far in a RNB target is 9kW (at Louvain-la-Neuve), and scheduled values at other facilities are summarized in Table 1 [5][†]. We note that the power given in this

Project, place	Particle	Energy (MeV)	Intensity (pμA)	Power (kW)	Target(s) Z	Fiss. rates (#/s)
KEK-JAERI, Japan	p	3000	333	1000	4-92	?
Rex-ISOLDE, CERN	p	1000	3	3	4-92	3×10^{12}
IRIS, Russia	p	1000	100	100	4-92	$\sim10^{14}$
SIRIUS/RAL, UK	p	800	100	80	4-92	$\sim10^{14}$
ISAC/TRIUMF, Canada	p	500	100	50	4-92	$\sim10^{14}$
GANIL(*), France	p or d	200	70	14	4-92	$\sim10^{14}$
Louvain-la-Neuve, Belgium	p	30	300	9	3-17	?

Table 1: Characteristics of RNB (ISOL) facilities in operation or projected, in which the direct production method is based on either p or d induced reactions.

Table is an incident beam power but not the beam power actually dissipated in the RNB target. Fission rates are estimated for 62.5g/cm^2 of UCx (ρ=2.5g/cm^3

[†]GANIL(*) estimates are from the case study reported in [7] as requested by the *Conseil Scientifique du GANIL.*

and 25cm long) except for the GANIL(*) project where 54g/cm^2 of molten U (ρ=18g/cm^3 and 3cm long) is taken.

The actual RNB target design problem can be viewed as one of minimizing target size and geometry while still handling the high beam power. Minimizing target size and geometry is essential to optimize diffusion and effusion efficiencies for the very short-lived radionuclides. In conventional ISOL production schemes (direct method), the target and ion source are integrally coupled so that the primary target can not be massively cooled without interfering with the ion source performance. With two-step schemes (converter method) these functions are physically separated. Table 2 lists a few examples based on this method [5, 6, 7].

Project, place	Particle	Energy (MeV)	Intensity (pμA)	Power (kW)	Target(s)	Fiss. rates (#/s)
ANL, USA	d	200	500	100	Be,W+U	5.5×10^{13}
LNL, Italy	p *or* d	40	2500	100	?+U	?
GANIL(*), France	d	35	5700	200	Li,C+U	1.0×10^{14}
ORELA/ORNL, USA	e	150	333	50	92	1.2×10^{13}
Photofission, Russia	e	25	20	0.5	W+U	1.5×10^{11}
MAFF/FRM-II, Germany	n	thermal	$10^{14}\frac{n}{s\,cm^2}$	3	^{235}U	1.0×10^{14}

Table 2: Characteristics of RNB (ISOL) projected facilities which are based on converter production method.

Finally, Table 3 presents a few examples of other projects based on a direct high energy projectile/target fission/fragmentation [5]. We note that both RIA

Project, place	Projectile	Energy (MeV/u)	Intensity (pμA)	Power (kW)	Target(s)	Fiss. rates (#/s)
RIA, USA	^{238}U	400	1/1	100	Li, C	$\sim10^{13}$
GSI, Germany	^{238}U	1000	1/6	40	Be, C	$\sim10^{12}$
GANIL(#), France	C	100	5	6	^{238}U	$\sim10^{12}$
GANIL(#), France	α	100	15	6	^{238}U	$\sim10^{13}$

Table 3: Characteristics of 3 typical RNB projected facilities which are based on direct projectile fission/fragmentation method. Note: GANIL actually operates with heavy ion beams up to 2kW. The beams of 6kW power and acceleration of α particles might be considered in the (near!?) future.

and GSI plan the slowing down of energetic fission fragments. If successful, these RNB factories will be able to provide intensive secondary beams both at high and low energies.

3 Comparison of the C- and D-methods

We assume that the main goal for the future RNB factory is to reach fission rates $\sim 10^{14}$fissions/s. If one comes back to the numbers presented in Tables 1- 3, $\sim 10^{14}$fissions/s might be considered "only" as a minimum requirement. Indeed, $\sim 10^{12}$–10^{13}fissions/s could be obtained even with present GANIL facility if 6kW beam power including acceleration of α-particles were allowed. We also take for granted that a powerful proton accelerator is available (say, protons of ∼1GeV and up to a few MW power). Tables 4 and 5 summarize the main beam and target characteristics within these two constraints for both converter (C) and direct (D) methods. In the case S1 (see Table 4), protons interact directly with

Scenario	Particle type	Energy	Min. primary beam power (current)	In target power	Target type (material)
D S1	p	500MeV	42kW (84μA)	16kW	solid UCx
C S2	p	500MeV	141kW (282μA)	7kW	solid W+UCx

Table 4: Projectile/target combinations to produce high fission yields defined by $\sim 10^{14}$fissions/s in the RNB production target. Also see Table 5. Note: C - Converter, D - Direct.

Target type	^{238}U density (g/cm^3)	Volume (cm^3)	^{238}U mass (g)	Geometry (cylinder)
D	2.5	177	443	"full": r=1.5cm, l=25cm
C	2.5	942	2355	"empty": r1(r2)=2cm(4cm), l=25cm

Table 5: Target parameters for the production of fission yields by the fission/spallation reactions. See Table 4 for details.

a solid UCx target (see line D of Table 5). In the case S2 (see Table 4), primary beam of protons impings upon a well-cooled W target to produce an intense flux of secondary neutrons. The secondary neutrons are of relatively low energy and nearly isotropic. To optimize the solid angle of the secondary UCx target, a cylindrical blanket is chosen (see line C of Table 5). The length and diameter of the secondary target are kept to a minimum for optimal extraction of short-lived products. Using thicker and longer secondary target can increase the yields of longer-lived isotopes.

The calculated in-target fission yields, normalized to a primary beam intensity resulting in 10^{14}fissions/s, are presented in Table 6 for both S1 and S2 scenarios. It is important to note that scenario S2 gives higher in-target fission yields on the neutron rich-side by a factor of ∼5 when normalized per successful fission. The yield of neutron-rich isotopes depends strongly on the excitation energy which is taken by neutron emission before and after fission. It is known that post-scission

Element	$T_{1/2}$ (s)	UCx (S1) (at./s 84μA p)	W+UCx (S2) (at./s 282μA p)
$^{91}_{36}$Kr	8.57	6.5×10^{11}	2.3×10^{12}
$^{94}_{36}$Kr	0.21	1.4×10^{11}	4.9×10^{11}
$^{80}_{37}$Rb	33.4	9.4×10^{9}	<1
$^{97}_{37}$Rb	0.17	7.4×10^{10}	1.5×10^{11}
$^{132}_{50}$Sn	40.0	1.9×10^{11}	1.1×10^{12}
$^{142}_{54}$Xe	1.22	1.5×10^{11}	9.0×10^{11}
$^{144}_{54}$Xe	1.10	1.1×10^{10}	7.0×10^{10}
$^{144}_{55}$Cs	1.02	2.2×10^{11}	1.1×10^{12}
$^{213}_{87}$Fr	34.6	6.7×10^{11}	6.3×10^{9}

Table 6: Estimate of projected in-target fission yields, normalized to a primary beam intensity resulting in 10^{14}fissions/s in the RNB production target. Also see Table 4.

neutrons are emitted by fission fragments with large neutron excess. That is why the fission of weakly excited nuclei (S2 scenario) is favourable for producing neutron-rich fission fragments. On the other hand, the S1 method will provide with higher intensities of certain spallation products (e.g. see $^{213}_{87}$Fr) and a broad range of neutron deficient nuclei (e.g. see $^{80}_{37}$Rb).

In both S1 and S2 scenarios examined we expect overal RNB source efficiency (release$\times$delay$\times$ion-source efficiency) similar to typical numbers given by ISOLDE target [8] due to similar target size and material compositions. This assumption gives $\sim$10% for some isotopes of the same element with $T_{1/2}$ >1s and around $\sim$1%-0.1% for $T_{1/2}$ <1s. In other words, the numbers in Table 6 have to be corrected accordingly for the expected final RNB intensities.

Due to $\sim10^{14}$fissions/s the total activity of about 10-20kCi is expected. Various parts of the installation will become highly radioactive. Hence, the management of this radioactivity is an important element in the design of the instrument. High radioactivity due to the noble gases and halogens will require a special treatment. Table 7 gives the maximum activity of the source after 90days of the irradiation. It is clear that this source will still be highly radioactive even

Cooling period	0s	1s	1min	1hour	1day	14days	30days	90days
Activity S1 (kCi)	11.6	10.7	8.3	4.8	2.6	1.0	0.7	0.3
Activity S2 (kCi)	17.7	16.6	11.5	6.2	3.3	1.0	0.6	0.2

Table 7: In-target activity of the UCx ion source after 90days of irradiation in the case of direct method (S1) and converter method (S2). See Tables 4-5 for irradiation conditions and target specifications for both scenarios.

after 90days of cooling. Consequently a remote control system will be needed to dismount a used source, and to mount its replacement.

We note separately that in the case S1 a number of long-lived α-radioactive spallation products will be produced on the proton rich side, which will define target activity in a long run (t=90days in Table 7). On the other hand, neutron induced fissions (case S2) will create more short-lived isotopes (see Table 6) on the neutron rich side resulting in higher target activity during operation (t=0s in Table 7).

4 Discussion

There are a few additional points to be discussed in the context as above. First of all, we think it is instructive to give the following example. The proton accelerator at PSI delivers 600MeV protons at 1MW power, i.e. 1.67p mA$\sim 10^{16}$p/s, which interacting with a massive Pb-Bi target surrounded by heavy water will result in $\sim 10^{17}$n/s over 4π. At the radial distance of 50cm from the spallation target the thermal neutron flux will be of the order of 3×10^{12}n/s/cm^2 to be compared with 1.5×10^{14}n/s/cm^2 as projected for MAFF/FRM-II (Germany) [5]. In other words, in the case of the PSI neutron source the ^{235}U (100%) mass has to be increased from 1g up to 50g to get 10^{14}fissions/s, or the primary beam power has to be increased by a factor of 50 (!?). In brief, the moderation of fast neutrons makes the powerful neutron source inefficient, simply because the RNB target requires a compact geometry and as little of fissionable material as possible. A reactor driven facility is strongly favoured in this particular case.

Secondly, if one thinks of the future RNB facility which is able do deliver not only intensive fission yields but also much broader range of isotopes, in our opinion, the p-driver could also provide it. For example, p(1GeV)+U would result in rather similar isotopic distributions as ^{12}C(100MeV/u)+U both on neutron-rich and deficient sides [8]. Thicker targets in the case of proton induced reactions (due to longer proton range) would simply compensate lower isotope production cross sections. In order to produce nuclei further on the proton-rich side of the mass valley, other reactions than nuclear fission should be explored. For this purpose lower mass targets than uranium or even very light mass targets should be used with 1GeV or higher energy protons [8].

This second point is extremely important in defining the production method (converter, direct, or both), which will directly depend on the main goals of the future RNB facility: delivering either the most intensive fission yields or being a broad range isotope production factory or both.

It has also been shown that thin Th targets ($\sim$1mg/cm^2) may stand very high beam intensities (>1mA) of high energy protons. A combination of a large number of such thin targets would allow eventually a chemically-independent production of RNBs (see [9] and Refs. therein for further details). Certainly, more R&D still has to be done in the case of this unconventional ISOL method.

5 Conclusions

The results we present show that very high fission yields may be obtained by the use of the powerful proton driver. For massive UCx targets ($\sim$2.3 kg of ^{238}U) the neutron induced fission from a converter can provide 10^{14}fissions/s with a primary beam intensity of 141kW for protons of 500MeV. The total power dissipated in the RNB target is $\sim$7kW. For less massive targets ($\sim$0.3 kg of ^{238}U), a direct p is preferred. 10^{14}fissions/s are reached with a proton beam of 42kW at 500MeV. The total power dissipated in the target is $\sim$16kW what is already very close to the present target technologies with allowable heat deposition due to the primary beam and fissions all together. It seems that the converter-method may reach the highest fission yields: 10^{15}fissions/s can be obtained employing the proton beam of 1.4MW at 500MeV, still compatible with the characteristics of a high intensity proton accelerator. On the other hand, in the case of the direct-method other targets than U could be used (e.g. Nb, La, Ta, Th or even much lighter mass targets) in order to explore different spallation/fission/fragmentation regions and proton-rich isotopes in particular. So the direct-method provides a higher versatility than the converter-method.

In brief, **the converter method** could provide **the highest fission yields** defined by **10^{15}fissions/s** in the RNB target. **A combination of the direct method** in addition to the converter method at the same time would produce **a broad range of other isotopes** of interest **at the intensities compatible with other projected facilities**. If all the suggested cases are technologically feasible is another important question that remains to be answered. Therefore, more detailed calculations and some exploratory experiments are necessary in this domain. Finally we add that the **radioactivity problems will be crucial** in the construction of the future RNB facility.

References

[1] W. Mittig, *J. of Phys.* ***G****: Nucl. Part. Phys.* **24** (1998) 1331.

[2] J.R.J. Bennett *et al.*, *Nucl. Instr. & Meth.* **B 126** (1997) 105; *Nucl. Instr. & Meth.* **B 126** (1997) 126.

[3] J.A. Nolen, Proc. of the 3rd Int. Conference on RNB, Gif-sur-Yvette, France, 24-27 May 1993, Ed. D.J. Morrissey, Editions Frontiers (1993) 111.

[4] D. Ridikas, "Optimisation of Beam and Target Combinations for Hybrid Reactor Systems and Radioactive Ion Beam Production by Fission", PhD thesis, report **GANIL-T 99 04** (October 1999) and Refs. therein.

[5] Book of Abstracts of the 5th Int. Conference on RNB-2000, Divonne, France, 3-8 April (2000); proceedings to be published in Nucl. Phys. A.

[6] J.A. Nolen *et al.*, Proc. of the 8th Int. Conference on Heavy Ion Accelerator Technology, ANL, USA, 5-9 October 1998, in press.

[7] D. Ridikas, W. Mittig, A.C.C. Villari, "Comparative study of the direct and converter methods for high fission yields", CEA report **DAPNIA/SPhN-00-51** (August 2000).

[8] H.L. Ravn *et al.*, *Nucl. Instr. & Meth.* **B 88** (1994) 441.

[9] W. Mittig *et al.*, *report of the TWIST workshop*, GANIL (2000).