SPIRAL Phase II European RTT Final Report

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SPIRAL PHASE-II

European RTT

Final report, September 2001

Contract number ERBFMGECT980100

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I Heavy neutron-rich beams induced by fast neutron induced fission

There is currently in the nuclear physics community a strong interest in the use of beams of accelerated radioactive ions. Although a quick glance at the nuclide chart immediately shows the vast unknown territories on the neutron-rich side of the valley of beta stability, only few of these projects are concerned with the neutron-rich nuclides. The SPIRAL-II RTD program aims at studying the techniques for delivering beams of neutron-rich radioactive nuclides at energies of a few MeV per nucleon. This energy allows overcoming the Coulomb repulsion between the radioactive beam and the target nuclei in most systems and opens up new possibilities for experimental studies of neutron-rich nuclei and of the synthesis of the heaviest elements. A number of new phenomena are indeed predicted to occur in nuclei with large neutron excess, which will help to improve nuclear models by comparison with data not available to date. Moreover, it should be noted that the astrophysics community is very interested in nuclear data for calculations of nucleosynthesis, which can lead to a better understanding of the observed abundancies of the various isotopes.

I-1 Principle

The technique originally proposed for SPIRAL-II is the use of energetic neutrons to induce fission of depleted uranium. The neutrons are generated by the break-up of deuterons in a thick target, the so-called converter, of sufficient thickness to prevent charged particles from escaping. The energetic forward-going neutrons impinge on a thick production target of fissionable material. The resulting fission products accumulate in the target, diffuse to the surface from which they evaporate, are ionised, mass-selected and finally post-accelerated. This method has several advantages. The highly activated converter can be kept at low temperature without affecting the neutron flux. Neutral projectiles then bombard the target losing energy only in useful nuclear interactions and having a high penetrating power which allows very thick targets to be used.

1-2 Methodology

This isotope separation on-line scheme produces intense beams of high quality. However, the proposed method requires further research on the following issues:

- Conditions to produce neutrons with high flux, narrow angular distribution and energy spectrum suitable for large fission cross-sections;
- Knowledge of nuclide cross sections as function of the incident neutron or deuteron energy;
- Estimates of the amount of nuclides produced in the target by coupling the neutron flux and the cross sections;

- Optimisation of suitable fissionable targets and of their efficiency and time constants for the release of fission products;

- Simulation of converter and target temperature distributions due to beam deposition and fission, which determines the maximum beam intensity allowed;

- Possibilities of producing a suitable deuteron beam with the present accelerator complex at GANIL or with another dedicated accelerator;

- The energy range of the radioactive beams which might be achieved, depending on the capacity to ionise and accelerate these elements with the available ion source technology and the existing accelerators at GANIL;

- Radiation safety aspects related to the neutron flux, activation of target material and other material as well as air in the experimental cave.

In the ISOL method several contradictory technical and physical constraints make it impossible to optimise freely each of the relevant parameters. The best values are therefore those, which allow best operation of the whole system taking into account the interactions between the different parts. For instance:

- Higher primary beam intensity increases nuclide production, yet it requires a larger target in order to evacuate the higher beam power deposition. However, diffusion of nuclides towards the target surface is the fastest for a small target. Thus, the extracted radioactive beam may not increase as much as expected. This applies especially to the most exotic and shortest-lived radioactive nuclides, but also to elements with long release times.

- Higher neutron energy opens new fission channels producing a vast range of nuclides and allowing a great variety of radioactive beams but it also favours the production of large amounts of unwanted activities that might be a problem with respect to radiation safety.

One of the main objectives of this R&D is to determine the intensity and energy of the primary deuteron beam giving the best yields of radioactive nuclides of interest for radioactive beams while taking into account beam power evacuation and safe operation of the facility. Our approach has consisted in carrying out simulations with various codes available or developed by our different task groups and performing a number of key experiments to validate the simulations. In this way, confidence is gained about the predictive power of the codes for situations where experiments could not be set up during the three years of this contract.

Evaluation of the potential of photo-induced fission in the same U targets as a possible low-cost limited scope alternative. This part of the investigation was added to the original proposal in course of the R&D programme.

1-3 Summary of the results

1-3-1 Neutron production

The concept of using neutrons generated by deuteron break-up implies a study of production yields, energy spectrum and angular distributions of neutrons in converters made of various materials and as a function of deuteron energy (see annex 1 for more details).

Experiments were performed at IPN-Orsay, KVI-Groningen and Saturne at Saclay. They explored a range between 14 and 200 MeV deuteron energy. The main features of neutron spectra are listed below:

- At forward angles, the energy distribution has a broad peak centred at about 0.4 times the deuteron energy. The angle of emission becomes narrower with increasing energy. For 100 MeV deuterons, the energy width (FWHM) of the neutron spectrum is about 30 MeV and the FWHM opening angle of the cone of emission is about 10 degrees.

- There is a rather isotropic distribution of neutrons of a few MeV due to evaporation in fusion reaction.

- The angular distributions and energy spectra are in fair agreement with calculations with an extended version of the Serber model and with the LAHET code. The Serber model reproduces the distributions of high-energy neutrons but not of the low-energy neutrons since evaporation is not implemented in the code. LAHET reproduces the low energy neutron spectrum while it tends to slightly underestimate (by less than a factor 2) the neutron distributions at very forward angles.

- A strong increase in neutron production is observed between 14 and 100 MeV deuteron energy. It is much less pronounced between 100 and 200 MeV. Among converters tested Be is slightly more productive than C. However, it has disadvantages related to its physical and chemical properties, as will be discussed later.

<u>1-3-2</u> Cross sections for n-rich nuclei in neutron-induced fission

Another step is the measurement of fission cross sections for n-rich nuclei at intermediate neutron energies. Experiments were performed at Jyväskylä in order to determine magnitudes and shapes of the distributions of nuclides in the mass range 88 to 144. These measurements combined mass yield measurements as a function of neutron energy (up to 50 MeV) and determination of charge distributions of isobars, measured by the IGISOL mass separation method (see annex 2 for more details).

Magnitudes and widths of distributions for neutrons of average energy 20 MeV generated by the 50 MeV deuteron beam on a ^{238}U target are similar to those obtained with 25 MeV protons. Yet, for a given element, they are shifted towards larger mass by about 2 mass units. In that respect, they are similar to the distributions of thermal n-induced fission on ^{235}U. However, fast neutrons produce a much wider distribution with higher cross sections at very asymmetric mass splits (A=80,160) and in addition, the dip in the symmetric region (A=120) is almost filled. These results have been used to extend a model originally designed for fission induced by intermediate energy protons.

Comparison with data for 2.5 MeV neutrons shows that the fission fragments emits about 1.4 neutrons more when the primary neutron energy is increased from 2.5 MeV to an average of 20 MeV. Due to increased excitation energy of the compound nucleus, high energy favours production of less n-rich fragments. On the other hand, the total fission cross-section increases up to about 40 MeV neutron energy and secondary neutron production increases with it. This implies the existence of a certain optimum neutron energy for producing n-rich nuclei.

<u>1-3-3</u> Studies with a converter + target system

It is also necessary to consider the geometry of the converter + target assembly. The target must intercept a large fraction of the neutrons. Thus, it must be close enough to the converter to subtend a large solid angle. The target temperature must be high enough to allow fast and efficient release of the fission products.

These studies have been performed with devices designed and constructed at the IPN-Orsay referred as PARRNe1 and PARRNe2 (see annex 3 for more details). They both include a converter and a target. Various materials for the converter and two different targets material, a porous UCx at high temperature and liquid uranium, have been tested.

PARRNe1 is a “portable” device, which can be installed in two weeks period by a five men team at another laboratory. It allows released yield measurements of gaseous elements. It has been used for measuring the isotopic distributions of Kr and Xe diffused out a thick uranium carbide target at IPN-Orsay, Louvain–la Neuve et KVI-Groningen. The target is able to stand the 2200° C maintained by an external oven. A gain in the production of noble gases by a factor larger than 10 has been observed when the deuteron energy increases from 20 MeV to 130 MeV. Most of the gain (a factor of 5) occurs between 20 and 80 MeV.
At GANIL, we have compared these yields (diffused out of the target) with the LAHET+MCNP+CINDER code predictions. The expected in-target yields are a factor 5-10 higher than the measured ones for the 80 and 130 MeV experiments. This difference is certainly due to the diffusion-effusion process. However, the expected in-target yields are in good agreement with the experimental ones at 50 MeV. We know, from the comparison of the experimental and simulated neutron angular distribution and of the Rb and Cs yields (from p (50 MeV)+U), that LAHET +MCNP+CINDER code underestimates the experimental data at lower energies.

This series of experiments concern only the production and diffusion of noble gases Kr and Xe out of the target. These gases are elements located in the region of asymmetric fission. The next step is to evaluate a wider range of nuclei in an ISOL system where fission products are delivered in the form of singly charged ions. PARRNe2 is an on-line mass separator constructed during this R&D and installed at the 15 MV IPN-Orsay tandem accelerator. It is a prototype for a feasibility study of the concept of SPIRAL-II, as well as a fully operational instrument for decay spectroscopy. Deuteron energies up to 27 MeV and intensities up to 600 nA are currently available.

In the most recent experiment at PARRNe2 with the fast-release UCx target (in December 2000) a MK5 plasma source similar to the one at CERN-ISOLDE has been used. PARRNe2 is able to deliver many of the elements available at ISOLDE. Among the very encouraging results is the measurement of the double magic ^{132}Sn produced, extracted from the target, singly ionised and mass-separated with a rate of 3.5 10^5/s. Extrapolation to 100 MeV and 500 µAe suggests that 1.7 10^9 singly-charged ions/s of ^{132}Sn can be delivered for charge breeding and post acceleration. The rates for elements extracted and ionized as singly charged ions are summarised in the annex 10.

Diffusion depends critically on the structure of the target material and on the temperature. At IPN-Orsay, R&D has been devoted to UCx targets following the technique elaborated at ISOLDE, and to molten uranium targets. The UCx and molten target are complementary. The higher temperature and shorter path to the surface in the grains of the porous UCx target favour fast diffusion ($T_{release}$ = 5 s for Xe). The liquid uranium target has a larger amount of uranium but slower release ($T_{release}$ = 112 s for Xe). The former is more efficient for short-lived isotopes, while the latter is more suited for longer-lived isotopes, with half-lives of about one minute. Further research is needed in order to find the optimum material structure, such as composition and density, for the targets and coupling to the ion source. These properties play a major role in the diffusion and thermal behaviour of the target.

The in-target yields can be calculated by various models and compared with measured mass-separated beam intensities. The ratio is the overall efficiency which is the product of efficiencies for release, transport to the ion-source, ionisation and transmission through the separator. In this way, release efficiencies can be deduced and cross-checked by internal consistency of the measurements. It should be noted that these values only apply to target + source configurations similar to those used in the PARRNe experiments. The lifetime dependence of the release efficiency has been deduced by comparing the experimental yields of radioactive beams with the estimated in-target productions. The in-target isotopic yields are available from the total fission cross-section from FICNeR code, adapted by the Orsay group to properly include the experimental geometry and from experimental isotopic yields already known for 14 MeV neutrons (from Los Alamos data bases). Knowing the ionisation and transfer efficiency, its possible to determine the release efficiency for each measured elements as done by the Orsay group with the experimental PARRNe2 measurements. A second approach, using the diffusion-effusion theory and the diffusion coefficients in carbon when available, has been done by the GANIL group. Good agreement is found for Kr and Xe radioactive beam intensities expected by the two approaches (see annex 10).

1-3-4 Choice of the deuteron energy

According to several experimental observations the deuteron energy of about 100 MeV is near the optimum value. This is discussed in more detail in annex 4. In short, this results from:

- The energy dependence of neutron yield, neutron energy spectrum and angular distribution as shown in annex 1.
- The energy dependence of total fission cross-section, which reaches a maximum at 40 MeV neutron energy.
- The energy dependence of nuclide cross-sections (annex 2 and annex 4): for a fixed number of total fissions, nuclei of the asymmetric region are better produced at lower energy while the others have a higher yields at higher energy. An energy of 100 MeV appears to be a good compromise.
- The experimental energy dependence of production rates for nuclides for the combined converter + target system, discussed in annex 3.
- The operation of the radioactive beam facility with respect to radiation doses and production of long-lived contaminants, discussed in annex 8.
- The LAHET and FICNeR codes also show that the number of fissions per kW of accelerated deuteron beam first strongly increases with energy but saturates near 100 MeV (see annex 4).

1-3-5 Power load on target and beam intensity limit

The maximum beam intensity which can be withstood by the converter or target define the maximum production rates achievable at the best deuteron energy:

- A general remark is that, since the beam power is proportional to the energy and the range roughly to its square, it is easier to dissipate a given beam power at high beam energy.

- The target temperature has to be high enough to enable quick diffusion of the fission products. It must also remain safely below the melting point of the target material. This results in only a fairly narrow range of operation, approximately between 2000 and 2200 C for UCx. The narrow temperature range justifies the choice of the converter method where no deuteron energy will be dissipated in the UCx target. The target is heated only by the stopping of fission products. This is not sufficient to maintain the minimum temperature of operation in the target. Extra heat has to be supplied by an external oven. This gives easy and safe control of target temperature.

Simulations of thermal behaviour have been performed at GANIL using the SYSTUS and Rubens codes at different beam energies and also different configurations with or without converter.

The converter can accept a beam intensity of 350 µAe at 100 MeV if the beam is defocused to a 2 cm radius. The limit is actually due to cooling of the outer surface of the C-converter. With a higher heat-exchange coefficient (but within a factor of 2) than is presently the GANIL standard, it should be possible to accept 500 µAe on the converter. An alternative solution, allowing higher primary beam intensity, would be a fast rotating converter. Such high power converters are in use at LAMPF and PSI and are being developed for the SPES (Legnaro) and R3B (GSI) projects.

The conventional method is to let the beam to impinge directly on the fission target. In this case, it is better to have the beam exits the target. This avoids unnecessary heating by the ions close to the end of their range (the Bragg peak), where the fission yield is neglegible. Nevertheless, only a 50 µAe beam of 2 cm radius is feasible for such a target. The major drawback is certainly the lack of temperature control independent of the beam intensity.

Yields have been obtained with the LAHER+MCNP+CINDER code for radioactive beam intensities consistent with temperatures which the target can stand, and an energy of 100 MeV. The 350 µAe beam on the converter induces 6. 10^{12} fissions/s in the target, while this is 1.2 10^{13} without converter with 50 µAe beam. However, yields of the most neutron-rich nuclei are higher with the converter, owing to the lower projectile energy (neutrons of 40 MeV average energy) and the formation of a more n-rich compound nucleus (no proton is captured).

The above-mentioned intensities imply a larger target than so far used at PARRNe. Diffusion out of such targets might be slower and reduce the efficiency for short-lived nuclei. Construction of large targets has been explored. The target used in PARNNe2 experiments is already suitable for deuteron beam intensities of 100 µAe at 100 MeV using the converter method.

More details about beam power dissipation and rates to be expected, with and without converter, are presented in annex 5.

1-3-6 Ionisation and post-acceleration

The atoms diffused out of the thick target must be ionised. The most recent PARRNe2 experiment, with a MK5 plasma source designed at ISOLDE, was promising. However, depending on the element to be ionised it is necessary to use several sources to guarantee the highest efficiency and/or selectivity. Considered so far are an ECR-source for gases and volatile elements, e.g. MONO1001 at GANIL, a surface-ionisation source for alkaline and earth-alkaline elements, and a more universal plasma source with a hot transfer line, e.g. the ISOLDE MK5 source. These sources could have efficiencies close to 100% for rare gases and alkalines, and between 10% and 50% for other elements. Moreover, a laser ion-source is well suited for specific cases where especially high purity is compulsory, but at the cost of lower efficiency.

Charge breeding is often considered necessary in order to accelerate the ions to final energies above the Coulomb barriers. Typically, the charge state should be increased up to at least one third of Z. Efficiencies are presently between 3% and 12% for the most probable charge, depending on the elements**.** Advances in this field are of crucial importance and the European RTD project contract “Charge Breeding” N HPRI-1999-50004 is being monitored carefully.

Several possibilities have been studied for post acceleration at GANIL. The cyclotron CIME allows acceleration of fission products to energies somewhat above the Coulomb barriers barrier for most projectile-target combinations. Coupling CIME and the second GANIL cyclotron CSS2 would allow to increase the energy in the range of 11 to 22 MeV/A. However, the necessary stripping of the beam before entering CCS2 would cause major beam intensity losses of about 80%. More details can be found in annex 6.

1-3-7 Deuteron driver accelerator

The case of an accelerator suitable for high intensity deuteron beams with an energy up to 200 MeV has been investigated at KVI and GANIL. For a short-term perspective the use of a cyclotron is the best option: it permits us to achieve the objectives of the project at the lowest costs. However, a consequence of this choice is that a significant increase in intensity in the framework of a future upgrade will require large additional investments.

Four options for the accelerator providing the deuteron beam for the neutron production in the SPIRAL-II project have been studied:

- The present GANIL cyclotrons. The radiation safety issuesalone of an 0.5 mA 80 MeV deuteron beam already exclude this option. For much lower intensities, this option might be useful but only in the commissioning of the target-ion-source assembly.

- The SARA booster-cyclotron, coupled to an injector. The maximum obtainable energy for this option is 72 MeV. For radiation safety reasons only stripping extraction of negative ions is viable. Also the injection and RF-system would require extensive refurbishing. Finally a 15 MeV injector (cyclotron) from which negative ions are extracted with a high efficiency (>95 %) has to be developed. Considering the cost and the rather complex operation of a coupled accelerator system this option is considered to be far from optimal but not technically impossible.

- A dedicated cyclotron. Very high extraction efficiency (>99.9 %) and high intensity (up to 1 mA) has so far been achieved with stripping extraction (IBA CYCLONE30 and EBBCO TR30) or with a large separated-sector cyclotron (PSI injector 2). The former solution is far more economic and easier in operation. It has shown its reliability in around 20 machines used for industrial radioisotope production. The development on the basis of the existing technology is straightforward and should allow a “turn-key” machine to be acquired. The stripping extraction makes it possible to vary the beam energy easily from the nominal value down to 50 % of this value. This option is therefore considered to be the optimal solution for the short term. More details can be found in annex 7.

- A dedicated linear accelerator. This kind of machine allows for significant increases in intensity and in energy within a context of future upgrade.

1-3-8 Safety considerations

There is, to date, no facility comparable to the design goals of SPIRAL-II. Therefore, radiation doses and production of contaminants have been estimated by using the LAHET-MCNP-CINDER code at GANIL. This has been carried out for different configurations, converter materials and deuteron energies. Considering the radiation doses (proportional to the beam intensity) for the same production of nuclei of interest, we arrived at the following conclusions:

- Alpha-activities are produced in large amounts at the highest energies since reaction channels for actinides are opened for more nuclei at higher energy. The α-activity is higher for a deuteron beam on a UCx target in the direct method than with a converter.

- Tritium production is larger with light-Z converters (C, Be or Li) than without. Carbon is the lowest producer of tritium among them.

For a fixed number of fission/s, the radiation doses during operation and most residual activities after a long-term shut down are weakly dependent on the energy. The only exception is for tritium production, which increases monotonically by a factor of 9 between 50 MeV and 200 MeV. Production of alpha-activities, 1 year after beam stopping, is clearly the lowest near 100 MeV. In conclusion, the energy of the deuteron beam for safe operation, taking into account the production of α emitters, is 80-100MeV. This is the same range as was determined to be best for production of neutron-rich nuclides.

Concrete shielding has been evaluated for the target-ion source vault and for a cyclotron driver accelerator assuming neutron production rates derived from LAHETcode.

Measurements have also been performed. The attenuation length of neutrons in concrete has been measured at GANIL in order to better estimate the amount of concrete shielding for neutrons needed during irradiation. Activation of air in the target area has been measured at Louvain-la-Neuve to validate the safety codes. Finally, a method has been developed and tested in order to measure the amount of tritium escaping from the target by diffusion. More details about safety aspects can be found in annex 8.

I-4 Proposals

Three proposals have been explored: a deuteron cyclotron, a deuteron linac, and photofission solutions.

I-4-1 Deuteron cyclotron solution

We examine the main characteristics of a proposal for the realisation of a radioactive beam facility in a rather short-term perspective (about half a decade) within the limits of existing technology. This is done in the context of a deuteron facility solution. A deuteron beam would be delivered by a new cyclotron at a maximum energy of 80-100 MeV depending on the cost. The energy can be reduced down to 40 – 50 MeV by adjusting the position of the extraction stripping foil. The converter method is proposed because of the safety of such operation. It is reasonable to assume that, after some additional research on heat transfer and cooling, a beam intensity of 500 µAe could be accepted by the converter + target system. The beam (or target) should be rotated and its width would need to be easily variable between 0.5 cm and 3 cm radius. The converter would be made of carbon. Its length is dictated by the range of the deuterons to be about 3.5 cm. Research is still needed concerning the nature of the carbon to be used as converter. It should guarantee a high thermal conductivity in order to keep an even distribution of temperature while it should minimise diffusion of tritium. Water or a more appropriate refrigerant liquid would cool the copper mechanical support. The maximum beam intensity delivered by the cyclotron should not be limited to 500 µAe in order to be able to increase the radioactive beam intensity if progress is made in improving the beam power dissipation in the converter in the future.

Two types of uranium carbide targets UCx would have to be made:

- A rather small target for 100 µAe deuterons when the shortest release times are required. A prototype (0.8 cm radius) already exists and has been used at the PARRNe2 on-line mass separator.

- A larger target (3 cm radius) for 350 µAe (or 500 µAe) deuteron intensity. This target could be made of 3 cm radius disks (if mechanically possible) or could be composed by combining 15 PARRNe targets. R&D is still necessary to determine thermal properties and diffusion characteristics of nuclides for various UCx compositions and densities.

An alternative possibility to use ThO_2 is also proposed, ThO2 being favorable for the release of certain elements, which are badly released from UC2/graphite.

An external oven using ohmic heating and with a surface temperature of 2200°C should be used to control and maintain the target temperature above 2000°C for efficient diffusion of fission products.

At least 4 types of 1+ ion sources will be necessary in the long term; an ECR type Mono1002 currently being developed at GANIL, a hot plasma MK5 from CERN, a surface ionisation source such as the Monolithe source also being developed at GANIL, and a laser ion source such as developed at IPN-Orsay. Modifications of MONO1002 and MK5 are required to replace the permanent magnets by coils to avoid loss of magnetic properties in the radioactive environment. These sources must all be adapted to the same outer dimension to allow exchange by a single robot. The vault should have two entrances for laser beams. Special care should be devoted to electric insulators concerning their high temperature properties and their electrical resistivity modification in presence of intense ionising radiation.

As technology progresses, the $1^+ \rightarrow N^+$ charge-breeding could be achieved either by a Phoenix ion source or by one of the new sources under development. In any case, the breeder source would need to be placed in a shielded area with a greatly reduced neutron flux outside the target cave.

In the early stage of operation of SPIRAL-II the CIME cyclotron will be able to accelerate radioactive ions up to and slightly above the Coulomb barrier for most cases. The expected “in-target” radioactive beam intensities are listed in annex 9. The expected radioactive beam intensities “after ionisation” are summarised in annex 10 for elements with rapid release.

This solution is based on in a rather short-term perspective (about half a decade) within the limits of existing technology. The rough price of such a solution (cyclotron, target-ion source, building and so on) is in the region of 28 MEuros, assuming that the cyclotron is obtained commercially.

I-4-2 Deuteron linac solution

In addition to fission, the deuteron solution outlined above would permit the rather difficult production of neutron-rich nuclei in the region above Z= 60 by reactions (n,p), (n,2p), (n,3p)…as is already done with the (p,n), (p,2n), (p,3n) reactions … for neutron-deficient nuclei. However, the above proposal (deuteron cyclotron) is a solution for a rather short-term perspective, with the possibility of accelerating deuterons with a limited energy range and without the possibility of upgrading the intensity above 2mA. With present technologies, only linear accelerators are capable to produce higher intensities. Such a linac solution has been investigated at GANIL. The first phase of such project is certainly much more expensive than other driver solutions; it also have greatly increases possibilities.

This type of machine could be constructed in two sections:

- A first intermediate step could allow, among several possibilities, deuterons of 40MeV, 5mA in order to get a total fission rate of 10^{13}/s, using a fast rotating converter as designed for the R3B project and at Legnaro (SPES);
- A further step, allowing a maximum energy of 100MeV/nuclon, with 300kW beam power for protons, deuterons, light ions with charge over mass ratio q/A =1/3.

Such a machine is a multi-beam driver, allowing to take benefit of the different types of mechanisms to produce the neutron deficient nuclei as the neutron rich nuclei. Moreover, it could also replace the role of the present GANIL machine for light ions with an intensity upgrade, which is not possible with the present GANIL accelerator. The second step is already a second-generation machine for the long term if we consider the potential radioactive beam intensities and the required converter-target and radioactivity management.

I-4-3 Photofission solution

An alternative to n-induced fission (this R&D) would be to use Bremsstrahlung – induced fission of uranium. In addition to this contract, the Orsay group, using the same PARRNe1 device and the same target, has performed measurement of Kr and Xe isotopic distribution produced by photofission and diffused out of a thick uranium carbide target. The LEP Preinjector at CERN has been used to deliver a 50MeV electron beam. The electron beam hits a thin tungsten converter of 4mm thickness. The bremstrahlung γ-rays irradiate the uranium carbide target. The measurements were performed with the 4mm tungsten converter in different positions (8 cm and 4 cm from the target) and also without the converter.
The Kr and Xe rates increase by an average factor of 1.5 when the converter is closer (4 cm) to the target. The gain is even larger (by about a factor 6) when the electron beam hits the uranium carbide target directly as compared with the converter placed at 8 cm from the target.
As identical uranium carbide targets were used and as the converter was placed at 8 cm from the uranium carbide for both electron and deuteron experiments, comparison is possible. Rough average yields factors (e/d) of 2.0, 0.5 and 0.3 were obtained between the experiments performed with 50 MeV electrons and with, respectively, 50, 80 and 130 MeV deuterons.

Considering the lower cost of an electron accelerator and the possibility of using an existing accelerator (MACSE from CEA-Saclay), this option is considered to be very attractive concerning the cost and the human resources. However, such a solution is far from optimal from the point of view of thermal dissipation and reliability: for a total fission rates of 10^{13}/s and a solution without converter, a power of nearly 20 kW (rough estimate), mainly due to bremsstrahlung, has to be dissipated in uranium carbide material for which the melting point is rather low. More precise power evaluations are in progress for different geometries, with and without converter. In the case of deuteron converter and for the same total fission rate, the power in uranium carbide target is nearly 0.3 kW, mainly due to fission products. The estimated price of such a solution is in the region of 19.5 Meuros, taking into account that a part of this accelerator already exists.

The plans for implementation at GANIL, are reported in annex 11 for these three proposals.

II Exploitation and dissemination of the work

The above-mentioned experiments have been analysed. Several publications have been accepted in refereed journals or are in preparation for submission. The results have been presented at several international conferences and various working group meetings. In addition, several PhD theses based on the project have been presented.

II.1 Publications in refereed journals

- F.Clapier, Phys. Rev.Special Topics - Accelerators and Beams, vol. 1 (1998)1-3.
- M.Mirea et al. Il Nuovo Cimento, vol. 111A, N.3 (1998) 267-279.
- S. Ménard et al., Phys. Rev. Spec. Topics AB **2** , 033501 (1999)
- D. Ridikas, W. Mittig and J.A. Tostevin, Phys. ReV. C **59**, 1555 (1999)
- S. Kandri-Rody et al., Nucl. Inst. Methods B **160**, 1 (2000)
- G. Lhersonneau et al., Eur. Phys. J. A**9**, 385 (2000)
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- D. Ridikas et al., Phys. Rev. C **63**, 014610 (2001)
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- V.R. Rubchenya and J. Äystö, Eur. Phys. J. A (2001), in press
- F. Ibrahim et al., submitted to European Physical Journal (2001)
- F. Ibrahim et al., in preparation (2001)

II.2 Conferences and meetings.

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II.5 Participants

This RTD contract has involved five European laboratories :

· the "Grand Accélérateur National d'Ions Lourds" (GANIL), Caen, France, coordinator of the project ;
· the "Centre de Recherches du Cyclotron" (CRC), Louvain-la-Neuve, Belgium ;
· the "Institut de Physique Nucléaire" (IPN), Orsay, France ;
· the Accelerator Laboratory (JYFL), Jyväskylä, Finland ;
· the "Kernfysisch Versneller Instituut" (KVI), Groningen, The Netherlands.

Furthermore, other experimental and theoretical groups, from Bucharest, CEA-DAPNIA, CERN, CSNSM-Orsay and St-Petersburg, although not directly involved in the present contract, have actively participated in the work.

III Summary

The results presented in section II above closely correspond to the project programme:

· The planned experiments have been carried out successfully. Codes for simulations of nuclide production, thermal behaviour of the converter and target and safety conditions have been written or improved.

· The best deuteron energy has been found to be in the range of 80 to 100 MeV. However, for a fixed number of total fission, nuclei in the asymmetric region are better produced at lower energy, while the others give better yields at higher energy.

· The converter method is definitely better than the direct method. A carbon converter is a good choice for the beam intensities required to reach the design goal of 10^{13} fissions / s.

· A prototype uranium-carbide target for intensities up to 100 µAe exists. However, R&D is still needed on feasibility of large UC targets and their diffusion properties. A R&D proposal on this critical issue has been submitted to the European Community, including European laboratories with interest in the ISOL method.

- A complete on-line mass separator PARRNE2 is now operating at the Orsay tandem accelerator as a prototype of the SPIRAL-II target + ion source assembly. Adaptation to higher beam intensities is nevertheless to be investigated.

- The various types of sources for ionisation of fission products as singly charged ions should cover the range of applications needed after modifications. The European RTD on charge breeding will be very beneficial for increasing the charge state of the ions before post-acceleration.

- The post accelerator CIME is suitable for acceleration of radioactive neutron-rich beams up to slightly above the Coulomb barrier. Injecting the CIME beam into the separated-sector cyclotron CSS2 will increase the beam energy to 11 – 22 MeV/A, however with substantial loss of intensity associated with the stripping before entering CSS2.

- The expected in-target radioactive yields for a 500 μA 100 MeV deuteron proposal are now available. The radioactive beam intensity (after acceleration) from a 500 μA 100 MeV deuteron facility have been evaluated for many neutron-rich isotopes.

- Concerning the deuteron driver, a cyclotron is a quick and less expensive solution but without versability andscope for evolution, whereas a linac solution allows a multi-beam driver. This last solution, despite of a higher cost, allows us to benefit from the different mechanisms to produce both neutron-deficient and neutron-rich nuclei. Moreover, it could also provide light and intermediate mass beams with intensities, which cannot be achieved by upgrading the present GANIL accelerators.

- Photofission is an alternative to n-induced fission (this R&D). In addition in this contract, measurement of Kr and Xe isotopic distribution produced by photofission and diffused out of a thick Ucx target were performed using the same PARRNe1 device and target. Comparison with the n-induced fission yields diffused out of the thick carbide uranium target are now available for neutron rich Kr and Xe isotopes.

Annex 1

1. Distributions of neutrons produced by deuterons on thick converters

Angular and energy distributions of neutrons are necessary to estimate production rates of n-rich nuclei. In addition, they provide data for dimensioning the shielding against fast neutrons. There existed in the literature only rather few data in the deuteron energy range between 14 MeV and 200 MeV. We therefore have performed a series of experiments as well as simulations in order to validate the computer codes.

1.1 Experiments

The neutron production has been measured as a function of neutron energy and emission angle for incident deuteron energies ranging from 20 to 200 MeV and for converters consisting of Be, C and depleted U (see table 1) [MEN99,PAU00a,PAU00b,PAU00c,RAD01]. The experiments have been performed at IPN (Orsay, France); LNS (Saclay, France) and KVI (Groningen, the Netherlands) and University of Jyväskylä (Finland). The neutron yields were obtained with a threshold of 2 - 4 MeV depending on the experiments. Some data for neutron production induced by low energy deuteron beams incident on thick Be- and C- targets have been published previously; our corresponding data are in good agreement with them.
Two processes at least contribute to neutron production. The first one includes the break-up of the deuteron during its collision with a nucleus in the converter and direct reactions where nucleons are knocked out of the nuclei in the converter. Neutrons produced in this way are emitted in the forward direction, where they dominate the neutron yield, and have a broad energy distribution centred at about half the energy of the incident deuterons. A second process, in which the deuterons are absorbed and neutrons are subsequently evaporated by the nuclei of the converter, produces mainly low-energy neutrons, which are emitted more or less isotropically.
The first process is the one of interest for the production of neutron-rich radioactive nuclides by fast neutron-induced fission. The strong forward peaking of the yield of high-energy neutrons (see fig. 1.1) shows that the approach with a compact geometry consisting of a converter to produce the neutrons followed by a second target containing the fissionable material is well suited to the task. Both processes have implications for the radiation safety of the proposed method. From these data and those obtained at lower energies the following main conclusions may be drawn:

- The neutron yield for all converters at first strongly increases with the incident energy and saturates above 100 MeV. This is illustrated in figure 1.2, which shows the neutron yield at 0° as a function of the incident energy of the deuterons for a Be-converter.

- The energy distribution of the neutrons produced in the deuteron break-up is centred at about 40% of the energy of the incident deuterons and has at 0° a width between one-third and half of the energy of the incident deuterons

- The width of the angular distribution of the neutrons produced in the deuteron break-up decreases with increasing incident energy of the deuterons. The FWHM is about to 21° at 17 MeV while it decreases down to 7° at 160 MeV.

- The absolute yield of high-energy neutrons from deuteron break-up on Be- and C-converters is significantly higher than for a U-c onverter. The yield of low-energy, isotropically emitted neutrons is significantly higher for U due to the additional source of deuteron-induced fission. This feature may be exploited for the production of specific radioactive nuclides having a higher yield in fission induced by low-energy neutrons.

- The neutron energy and angular distributions are similar for Be- and C- converters. The yield for a Be converter is about 30% higher and it is slightly more forward peaked than for a C-converter.

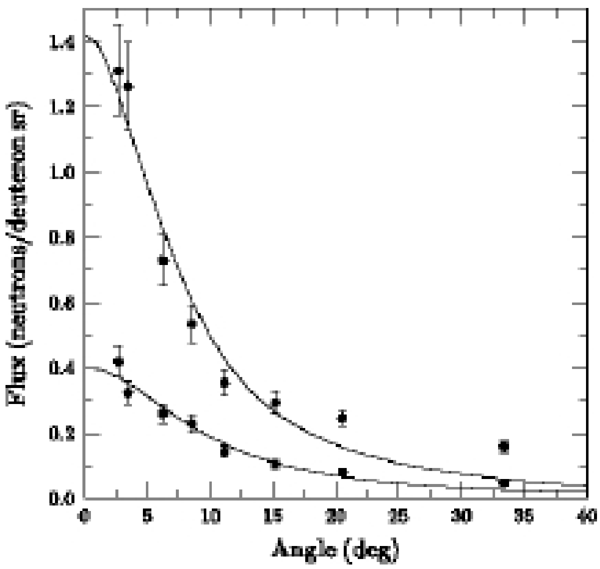

Fig. 1.1: Experimental angular distributions of neutrons for 80 and 160 MeV deuterons incident on a thick Be target.

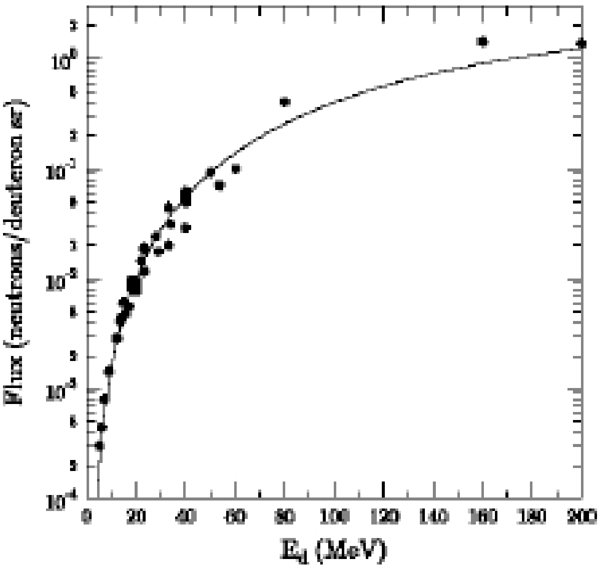

Fig. 1.2: Neutron yield at 0° as a function of the incident deuteron energy for a Be-converter.

1.2 Comparison with the Serber model

In order to allow modelling of the proposed method a theoretical model of the neutron yield has been developed by the Orsay group in the framework of the model designed by Serber [MIR00]. The curves in figure 1.1 represent the predictions of this model, which are in satisfactory agreement with the data. The first component of the model has one normalisation factor adjusted to the neutron yield at 0° as a function of the incident energy of the deuteron. The second component, extending the Serber model [SER47], introduces another adjustable parameter to account for direct nuclear reactions knocking neutrons out the target nucleus. The model does not take yet into account the low-energy neutrons with isotropic angular distribution produced by the evaporation process nor, in case of a U converter, by fission. The calculation consequently underestimates the yields at the largest angles.

1.3 Comparison with the LAHET code

Simulations of neutron spectra have been performed with the LAHET high-energy transport code combined with the Monte Carlo N-Particle MCNP code for low-energy transport. Deuteron Coulomb dissociation has been added to the more standard processes (the forward peaked break -up and direct reactions and the rather isotropic evaporation of low-energy neutrons) [RID99]. The simulations performed at GANIL [SAI01] are compared with experimental angular distributions measured at energies of 80, 160 and 200 MeV for C and Be converters. Comparison with experiment (80 MeV d + C) is shown in figure 1.3. The LAHET-MCNP code reproduces the order of magnitude of the differential cross sections without any adjusted parameter. However, some systematic deviations can be noted. At 80 and 160 MeV, the intensity of the distributions is underestimated at the very forward angles only. The deviation is the most pronounced as the deuteron energy decreases. In contrast, at 200 MeV LAHET overestimates the productions especially at the small angles and the calculated mean energy is too low. This difference could be a consequence of systematic errors due to the different experimental methods used (time of flight at 160 MeV and activation at 200 MeV) .

1.4 Outlook

The neutron yield is only one of the factors to be taken in consideration for the choice of the converter material. Other aspects in the evaluation are:

- thermal properties in view of a compact geometry of the converter and production target ;
- toxicity and material properties of the converter;
- production of long-lived radioactive nuclides;
- cost of operation.

Conclusions regarding are these aspects are summarised below:

- A Be converter produces the largest amount of neutrons. However, the low melting point (1278° C) does not allow high-intensity deuteron beams nor its placement very close to the hot target.
- Liquid lithium is a more robust converter with respect to deposited beam power than Be or C. However, the flow of a hot Li-liquid containing some amount of radioactive products requires special care of design especially due to safety considerations. A converter designed along the lines originally described by Grand and Goland [GRA77] is probably not to be considered in the context of SPIRAL-II which is a radioactive beam facility of the first generation, but could be of interest for a next generation facility, e.g. EURISOL since it can stand very high beam intensity.
- The above mentioned properties clearly favour graphite as converter material. It is non-toxic, easy to handle and has a high melting point of 3632 °C. These excellent properties allow for high beam intensities with only simple water cooling on the outer surface (see annex 5). For a second generation machine, it could be envisaged to use the same converter cooled by He gas as first reported by Talbert et al. [TAL92].
- A UCx-converter would allow an even more compact geometry in which the converter and production target are completely integrated. The performance of such a set-up would be

improved with respect to the reference case with a C- converter in two ways. Firstly, the neutron flux in the production target is increased because of the geometry. Secondly, fission in the production target results not only from neutrons from the deuteron break-up but also from a few other sources like protons from the deuteron break-up, incident deuterons and secondary neutrons from fission of U. However, the melting point of UCx, about 2400 C, limits the beam intensity while fast diffusion requires temperatures above 2000 C. This leaves only a narrow range of temperature for safe and efficient operation. It must be noted that the heat distribution will be highly non-homogenous, leading to local overheating (where the beam is stopped) whereas colder regions would be acting as traps for the fission products. A solution with a thinner target where the deuteron beam is not stopped was also investigated.

For the cases mentioned above the maximum allowed beam intensity and fission rates have been calculated, (see annex 5). Another possibility is to distribute the heat by using a rapidly rotating converter. This solution is applied by LAMPF at Los Alamos (USA) and PSI [Hei00] at Villigen (Switzerland) and is also being studied in the context of the R3B RTD project.

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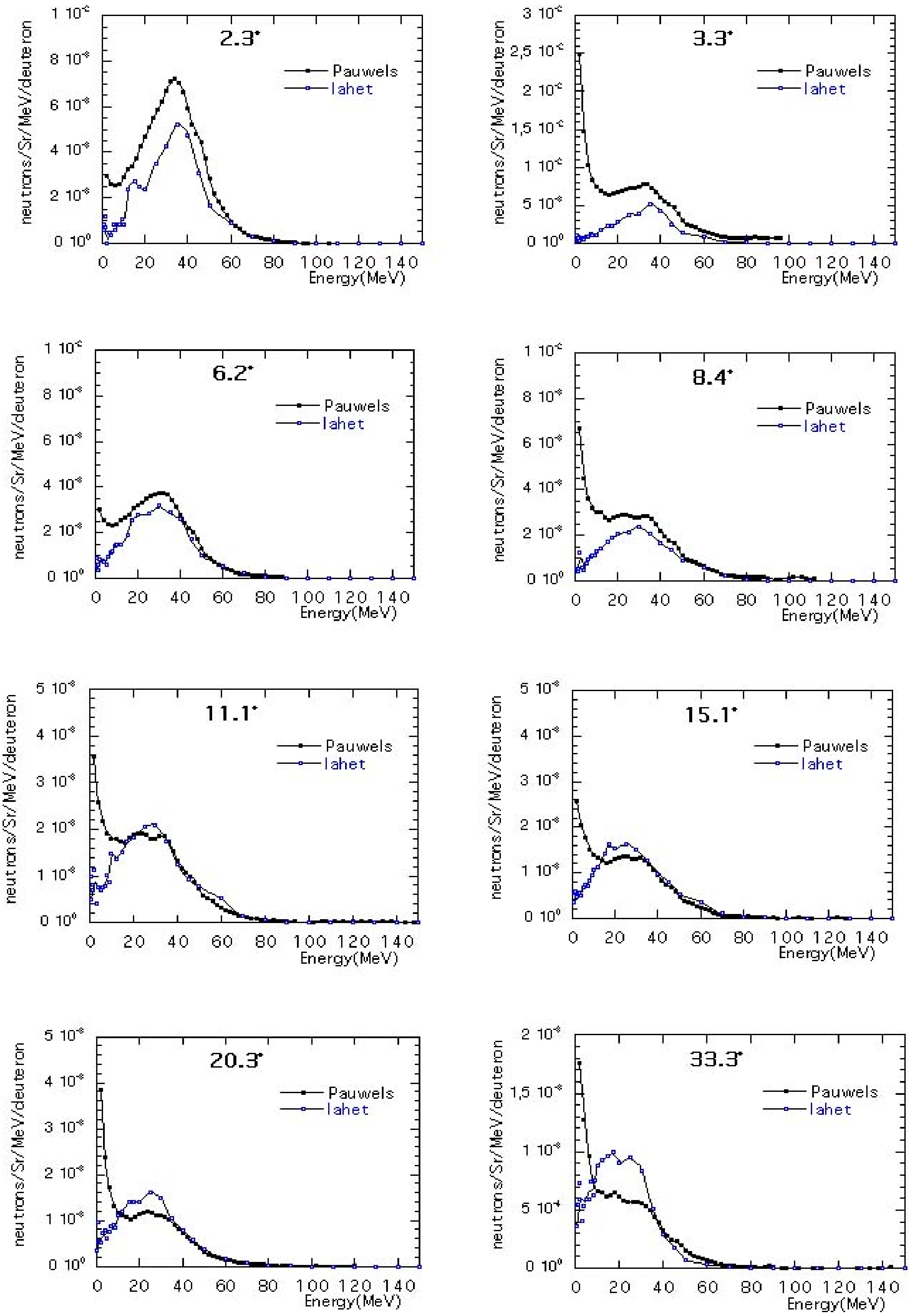

Fig.1.3 : Experimental and simulated differential cross sections of neutrons produced by 80 MeV deuterons incident on a thick C target.

Annex 2

2. Cross sections for nuclide production

Knowledge of cross section dependence on incident projectile energy is necessary to estimate production rates of n-rich nuclei. It is especially crucial to determine the gain in cross section when using fast neutrons or deuterons on ^{238}U with respect to thermal neutrons on ^{235}U or protons of intermediate energy on ^{238}U. In the energy range between a few and 200 MeV, fission is the main mechanism for production of neutron-rich nuclides. After a literature survey, we have performed a series of measurements in order to extract cross sections for nuclides with neutrons of 20 MeV average energy generated by a 50 MeV deuteron beam [LHE00]. As it was impossible to perform several such time-consuming experiments at various energies during the time span of this R&D, computer codes have also been used. Their validity had to be tested from comparisons of their predictions with experimental results for proton-induced fission where more data are available. The following codes could be applied :

- The LAHET+MCNP code used for neutron flux simulations has been combined with the CINDER code. This work was done at GANIL for the purpose of SPIRAL-II [RID99]. The combined code allows calculations of yields of fission products, integrating the geometrical conditions of converter and target. LAHET also includes other reaction mechanisms such as target fragmentation, spallation and transfer. The latter plays a role in production of actinides and is consequently of importance for radiation safety. However the code, originally designed for a high energy regime, could possibly not be well suited to the low energies considered here.

- A code especially designed to calculate cross sections for intermediate energy fission at Jyväskylä by V. Rubchenya. This code (we refer to it later as VR) is continuously being upgraded to take into account new data on p-induced fission obtained at the IGISOL facility where the proton energy is typically 25 MeV. It includes also a fission mode, so-called super-asymmetric, which enhances the yield of neutron-rich nuclei in the region of A=80 [HUH97]. Boosted by recent works, e.g. SPIRAL-II and the Dubna project, the code has been extended to n-induced fission and photofission. However, predictions of production rates in the converter + target geometry (which integrates the actual neutron spectrum impinging on the target) are not available. Mechanisms other than fission are not included. This limits the use to low energy regime.

- A new code integrating neutron production in the converter and neutron-induced fission in the target (FICNeR), taking into account the geometry of the assembly, has been designed during the course of this project [MIR00]. The code is well-suited to model experiments with PARRNe discussed in annex 3. The acronym FICNeR stands for fission induced in a target by fast neutrons. The model consists of two parts. The first one describes the fission process as function of neutron energy using a geometrical approach of the shape of the nucleus. The second one combines the data on neutron energy and angular distributions and the geometry of the converter and target system to calculate production yields for nuclides in the specified geometry. Fission cross sections have two contributions of comparable magnitudes (0.5 b). Fission of ^{239}U starts at about 1 MeV neutron energy whereas fission of ^{238}U created after evaporation of one pre-scission neutron starts above 6 MeV. However, other processes than fission by the incident neutrons such as transfer and fission by second generation neutrons are not taken into account .

2.1 Literature survey

There are comparatively few experimental data of relevant for the production of neutron-rich nuclides induced by fast neutrons:

- Thermal neutrons: the conventional method has long been fission of ^{235}U by thermal neutrons using a high-flux reactor. This method was chosen by the PIAFE project at the ILL-Grenoble [VIG96] now revived as MAFF at Munich [MAF00].

- At 2.5 MeV: Philips et al. looked at gamma-rays emitted in the fission of ^{238}U by neutrons produced by ^{7}Li + H [PHI98]. A few isotopic yields are reported for elements in the Zr and Ba region.

- Up to 14 MeV: Cumulative yields of isotopes in fission of ^{232}Th and ^{238}U have been investigated using reactor neutrons or neutrons produced by reactions. Radiochemistry was used to isolate and identify long-lived fission products [RAO74, NAG78, RAO79]. An enhancement of cross section on the wings of the mass distribution (A about 80 and 160) was observed with respect to thermal neutrons.

- At higher neutron energy: no cross sections for nuclide production are available. Nevertheless, mass distributions were measured for fission of ^{238}U by neutrons of energy from a few MeV up to 800 MeV [ZOE95]. A comprehensive experiment is planned at Los Alamos in order to determine isotopic cross sections by looking at gamma-rays [ETH01].

- Let us also mention the comprehensive study published during the course of our experiments by the Studsvik group with reactor neutrons on ^{233}U [GAL00].

More experimental data are available for production of nuclides by protons. The most systematic studies being carried out with:

- 25 MeV protons with the IGISOL technique at Jyväskylä [JAU94, HUH97]. Other light projectiles, like deuterons [LEI91] and alphas [AST92] were used occasionally;

- 50 and 150 MeV protons at the Orsay synchrocyclotron [CHA70];

- 600 MeV or 1 GeV protons at CERN-ISOLDE [RAV94];

- Inverse kinematics with 1 GeV per nucleon ^{238}U on hydrogen at GSI-Darmstadt [SCH01].

Cross sections in thermal-n-induced fission are several orders of magnitude larger than with charged particles in the regions of asymmetric fission (i.e. A=95, 145). However, yields are very low in the region of symmetric fission (A=120) and the use of a reactor is bound by severe safety constrains. In contrast, the higher energy of charged particles creates more evenly distributed mass splits. This feature is of advantage regarding the diversity of beams delivered by the future facility. The same is expected from fast neutrons generated by break-up of the deuterons.

2.2 Experimental cross sections for neutron-induced fission using 50 MeV deuterons on a C-converter

A dedicated measurement of cross sections has been performed for 50 MeV deuterons on a C-converter at Jyväskylä The measurements were performed in several steps. First, mass-yield measurements were carried out for various neutron energies by using the (d,pf) reaction. Then, with well defined geometry of the C-converter the neutron angular and energy distributions were determined. Finally, Z-distributions for selected masses were measured under the same neutron flux conditions as determined in the second step with the C-converter. The combination of these data subsequently yields cross sections for individual nuclides.

2.2.1 Mass distributions as function of deuteron energy

Fission total cross sections are available in the literature for a range covering a few MeV to 300 MeV [LES94,EIS96]. An experiment in collaboration with the HENDES group was performed in May 1998 to determine the distributions of the fission fragment masses as a function of the neutron energy up to 50 MeV. The 65 MeV deuteron beam bombarded a thin natural uranium target. The protons emerging after dissociation of the deuteron and the fission fragments were detected. Measured velocities, angles of emission and energies of all ejectiles enabled reconstruction of the neutron energy and of mass splits using momentum and energy conservation [RUB00,TRZ00]. The energy dependence of mass distributions is shown in Fig. 2.1. This measurement is in good agreement with another one performed at Los Alamos where high-energy neutron beams are available [ZOE95].

2.2.2 Neutron energy and angular distributions for 50 MeV deuterons on C-converter

In order to know the exact characteristics of the neutron flux impinging on the uranium target two experiments were performed. The first one was also carried out by the HENDES Collaboration for Be- and C- converters [RAD01]. The angular and energy distributions were measured with large position-sensitive liquid scintillators, and by time of flight. They were found to be in good agreement with those measured by Meulders et al. [MEU75]. The second one was a cross- checking of the experimental conditions for the next series of measurements by on-line mass separation with the same converter and target geometry. It was made by measuring the activation of metallic foils in placed of the uranium target. About 3 neutrons were produced per 100 deuterons incident on the C-converter, of which 40% actually impinged on the target.

2.2.3 Charge distributions in selected mass chains

During a last experiment isotopic distributions were measured using the IGISOL facility at Jyväskylä. This on-line mass separation technique allows fast extraction, thus without decay losses, of the fission products and with a negligible chemical dependence of the efficiency. A special stopping chamber has been constructed in order to take full advantage of the absence of ‘plasma effect’ in the case of neutron beam. The production rates of neutron-rich short-lived nuclei in the

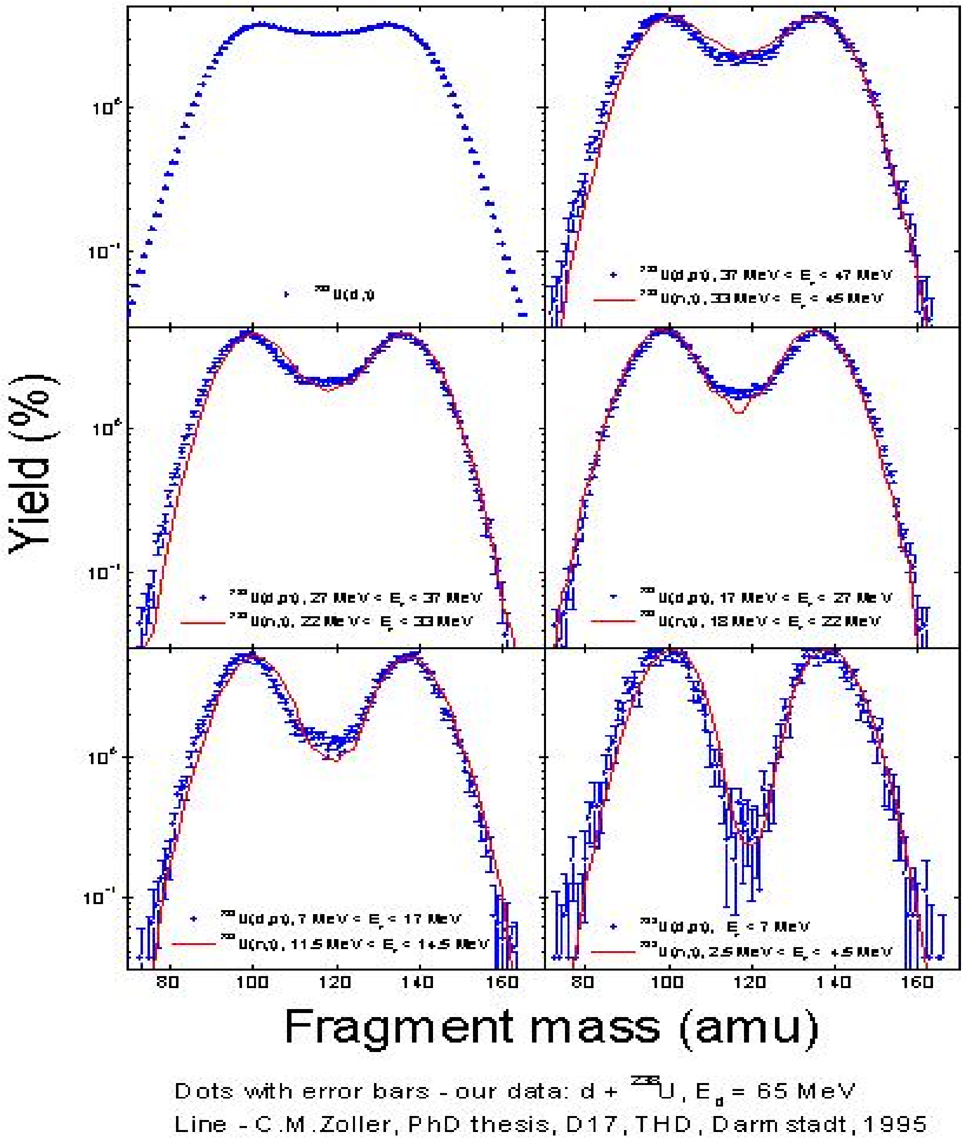

Fig. 2.1: Yields versus fragment mass. Dots (with error bars) are the data from our experiment with 65 MeV deuterons, obtained by kinematic reconstruction based on energy and time measurements of proton and fission products (the neutron absorbed in the target after breakup of the deuteron is not detected directly). The solid line represents the data from C.M. Zöller [ZOE95] where the neutron energy was measured by time of flight. There is good agreement in spite of the different methods. The fact that the our mass resolution was a few units and the data not unfolded can account for some widening on the wings. The top leftmost graph is for complete fusion, otherwise for neutron energy bins as indicated.

mass ranges 88-94 and 136-144 have been measured. The data were normalised so that the sum of nuclide cross sections within a mass chain adds up to the mass cross section (see § 2.2.1). Among these nuclei are the Kr and Xe isotopes, which are also produced and diffused out of thick targets and therefore of special interest for monitoring the design parameters of the converter + target. A similar measurement has been performed with 25 MeV protons on uranium in order to compare the cross-section distributions in magnitude, width and location of the maximum in the (Z,A) plane for these reactions. The data allow one to calculate fission cross sections and the production of fission fragments in the target for a wider range than was actually measured. This can be done using a simple empirical interpolation [LHE00] or the more physical model (VR) developed by V. Rubchenya adjusted to take into account these new data.

2.2.4 Comparison between spontaneous, n and p-induced fission experimental data

In spite of weak statistics, one can extract some conclusions based on clear trends exhibited by the data:

- Data for 20 MeV neutrons on ^{238}U show that the fission valley is almost filled, which is not the case with thermal neutrons on ^{235}U. Within isobaric chains the charge distributions are Gaussian with σ =0.7 charge units.

- The magnitudes and widths of the cross-sections appear to be roughly the same as for fission by 25 MeV protons, but there is a shift of the centre of the isotopic distributions. This shift is typically of 2 amu , favouring production of neutron-rich nuclei with respect to proton-induced fission. Fig. 2.2 shows relative positions of the maximum of distributions for various production modes.

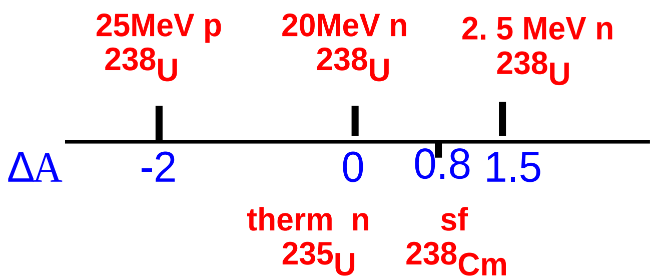

Fig. 2.2: Relative shifts of the mean value of isotopic distribution for elements in the range Z=34-56 for various methods discussed in the text. The reference value is given by the empirical equation A_p = 2.730 Z –8.8 reproducing data for 20 MeV average energy neutrons on ^{238}U [LHE00].

2.3 Validation of the codes for proton and neutron-induced reaction

Except for the experiment described above there has been no nuclide independent cross-section measurements for fission by fast neutrons. Therefore we start by comparing the calculations with experiments with proton beams, then with the experiment with neutrons described above and finally give some predictions for neutron-induced fission.

We first compare the results of the VR code with the experimental cross-sections for 25 MeV protons. The data are routinely used to improve the code. Thus, the excellent agreement is not surprising. The implementation of the super-asymmetric channel, necessary to reproduce cross sections in the A=80 region, seems to be somewhat too optimistic regarding production of the most exotic nuclei. The calculated most probable A(Z) values are a bit too high according the most recent measurement at the LISOL separator [FRA98].

The comparison with the experimental cross-sections for fission induced by about 20 MeV neutrons is also satisfactory. The calculation confirms a shift of the maxima of the isotopic cross sections in p and n-induced fission. The average value of 1.2 amu is somewhat lower than the experimental value of 2 amu.

A comparison of the LAHET and the VR predictions can be done with experimental cross-sections for Rb in fission induced by 50 MeV protons. The data have been obtained by the ISOL on-line mass separation using a target with a very fast release time $T_{release}$ < 300 ms [CHA70]. The VR code is correct in magnitude but overestimates the production of the extreme n-rich nuclei. The standard option in LAHET produces curves which are too broad for the cross section of isotopes of a given element. The inclusion of a pre-equilibrium model allows charge and energy to be released before giving a chance to fission [RID99]. LAHET is correct in the position and width of the isotopic distributions but underestimates the yields by about a factor of 3 [SAI01].

Finally, we compare some predictions of the models. In general, LAHET predicts a smaller cross section than the VR code. The largest differences (fby a factor of about 7) are observed especially for masses near A=80 (super-asymmetric region) and near A=115 (symmetric region). The larger cross section by VR is obviously due to the explicit implementation of a symmetric and a super-asymmetric mode. Yet, predictions are quite similar for the isotopes on the n-rich side in the A=90 and 140 asymmetric regions.
The nuclei ^{78}Ni and ^{132}Sn are of special importance due to their nuclear properties (large binding energy for these so-called doubly-magic nuclei) which makes them well suited as beams for producing heavy elements with low excitation energy. As stated above, VR predicts a larger cross-section for ^{78}Ni. For n-rich Sn isotopes, it predicts a strong decrease of cross sections with increasing energy whereas the dependence is very weak with LAHET. Validating the codes would require dedicated experiments as a function of neutron energy and other measurements in the low yield region on the wings of the distributions. These are extremely time consuming with the present experimental conditions.

2.4 Outlook

There is still a need for further experiments to give confidence to model predictions in a wide range of masses and energies. So far, the experimental facts of relevance can be summarised as follows.

- Thermal-neutron induced fission of ^{235}U has the highest cross sections in regions of asymmetric fission but has a very deep valley in the symmetric region and falls off very fast on the low A =80 (and high A=160) region. The environment of a high-flux reactor and the need for a fissile target imposes severe constraints.

- Low energy (e.g. 2.5 MeV) neutron-induced fission of ^{238}U produces the most exotic isotope distribution. However, cross sections are low and the envelope of mass distribution is not yet very different from the one for thermal-neutron induced fission.

- Intermediate energy (e.g. 20 MeV) neutron-induced fission of ^{238}U is interesting for production due to the overall increase of cross section compared with 2.5 MeV and further increases in the symmetric region and on the edges of the mass distribution due to the opening of new fission modes. Compared with 25 MeV proton induced fission, cross sections are similar in width and magnitude, but production of n-rich isotopes is favoured by the shift of the maxima of cross sections to higher N for a given Z.

- Further increase of the energy will move the distributions closer to stability. However, with increasing deuteron energy the neutron production per deuteron increases and the forward focusing of the neutrons improves, whereas the solid angle intercepted by the target decreases due to the increased converter length. After 100 MeV deuteron energy the neutron production tends to saturate. This energy, corresponding to the maximum of the fission cross section (at 40 MeV neutron energy) must be close to optimum. It has to be investigated if it is compatible with technological constrains of the design of converter and target assembly. This is done in annex 5.

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Annex 3

3. Target development and realisation of radioactive beams

So far, we have studied the neutron flux and neutron-induced fission cross sections. They allow us to calculate the in-target production rates. The radioactive beam intensity is directly proportional to the amount of fission product extracted from the target. This part deals with target development and the performance to extract (in the first instance) gaseous elements as function of deuteron energy and nature of the converter and then to produce low-energy beams of radioactive ions for a wider variety of elements. For that purpose, uranium carbide and liquid uranium targets were developed. Then, a set-up to measure gases diffused out of the targets (PARRNe1), and a complete dedicated on-line mass separator (PARRNe2) were constructed.

3.1 Target development

Development of target technology is necessary to increase the release efficiency and to shorten the time the fission products spend in the target material (release time) for the largest possible number of elements. These properties are vital for making intense beams of short -ived nuclei.
The release of products created inside the target is governed mainly by three parameters: the temperature which favours diffusion, the target thickness and structure since a shorter path in the matter favours the release, and finally the desorption at the surface of the target. These effects favour a porous and high temperature solid over a liquid. However, the amount of material is definitely larger in a liquid target. For a UCx target containing 33 g, a molten target in a comparable target volume contains 250 g. Therefore research has been continued towards both a high temperature solid uranium carbide target UCx with the fastest release times, and a high density molten target with the largest amount of fissile material.

3.1.1 Uranium carbide target

The release of fission products depends essentially on diffusion in the grains and desorption at the surface of the grains. Diffusion depends on a parameter μ with the dimension of a decay constant and inversely proportional to the square of the grain radius. Moreover, μ and the sticking time at the surface strongly depend on temperature with an exponential dependence on 1/T. Release is thus favoured by a small grain size and high temperature.
A UCx target has been prepared following a carburation technique routinely used at CERN-ISOLDE. It consists of pellets made of grains of 44 µm placed in a graphite container and heated by a cylindrical C-oven. Various targets with different C:U proportions and densities have been developed. A first target with 33 g of ^{238}U was tested at 2000C and used at IPN-Orsay, Louvain-la-Neuve and KVI with PARRNe1. The main parameters were: a U-density of 2.3g/cm^2, a length of 50 mm, a radius of 14 mm, and a ratio of 9 carbon atoms for 1 U atom. The target has been improved with a length of 150 mm and a ratio of about 1.7 carbon atoms for 1 U atom. It has been tested up to 2200°C at the IPN-Orsay with PARRNe2.

3.1.2 Liquid uranium target

A liquid target is a priori attractive. Self-diffusion coefficients in a liquid are higher than in a solid by 2-3 orders of magnitude, convection favours the transport of radioactive isotopes to the surface and the target can contain much more material in a compact geometry. However, the

path to reach the surface is considerably longer than in a solid target made of grains and the surface layer is a barrier that can suppress the outgoing flux. The temperature is limited by the evaporation of the target and the chemical reactivity of uranium. In practice the crossing of the surface determines the release time.
A 20 g liquid target was investigated with PARRNe1 at IPN-Orsay [KAN00]. In the most recent on-line experiments (May 2000) with PARRNe2 a molten target with 250 g of uranium was used. It was placed in a cylindrical Y_2O_3 ceramic container surrounded by a Mo container forming a barrier in case of a leak. The whole was placed in the graphite oven. The target withstands a temperature of 1600°C for several days and it has been tested so far with success at 1700°C.

3.1.3 Uranium carbide versus liquid uranium

A comparison of the performance of both targets at PARRNe2 is shown in Fig. 3.1. For isotopes having lifetimes longer than 1 hour, the production rates with liquid uranium are higher by a factor of about 2-3 which is below the gain of factor 8 due to the increases of uranium atoms. The liquid target, due to its slower release, looses efficiency. For nuclei with the shortest lifetimes, uranium carbide target is even better.

It has been observed that at 1700C the uranium had migrated along the walls of its container. Thus, there might be room for further improvement with the molten target with respect to better containment as well as for the transfer of fission products to the ion source in the present design.

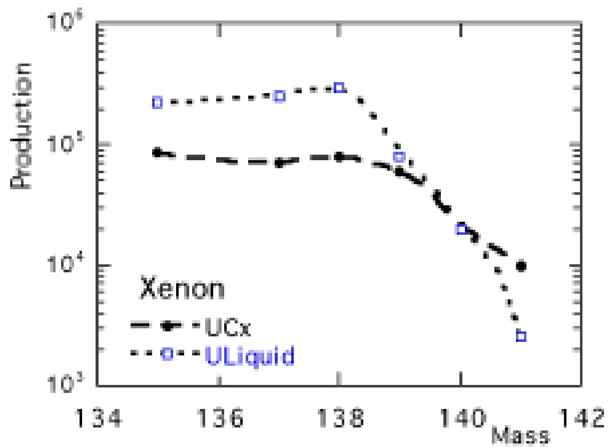

Figure 3.1: Comparison of yields of Kr isotopes collected as 1^+ ions in the focal plane of the PARRNe2 separator for 30 g UCx and 250 g molten uranium targets, respectively.

The considerations discussed above suggest that the UCx and molten target are complementary. The UCx target has faster release but less uranium than the molten target. Thus UCx is better suited for the very n-rich fission products and isomeric states while the molten target is interesting for high production rates of moderately exotic nuclei.

3.2 Production of gaseous elements

3.2.1 Experiments with PARRNe1

Gaseous elements have been chosen to evaluate quantitatively the production rates as function of various design parameters. Noble gases diffuse at high temperature without the necessity of an ion source. This allows experiments with a portable measurement set-up PARRNe1. Experiments at various laboratories participating in the R&D provide a variety of deuteron beam energies and intensities delivered by their accelerators.

The device PARRNe1, built by the IPN Orsay (IN2P3), has been used for measuring the isotopic distributions of neutron-rich isotopes of Kr and Xe. Fission fragments are produced by irradiation of the target by neutrons generated in a converter. They are slowed down, stopped and become neutral atoms inside the thick target. Only the gaseous elements are able to diffuse out of the target, to be transferred through a cold tube of several meters and be collected on a cryogenic cold finger. Identification is made by γ-spectroscopy. The production of n-rich Kr and Xe isotopes has been measured as a function of the following parameters: temperature of the target, geometry of the converter and fission target and incident deuteron energy.

The influence of the energy of the deuteron beam has been studied by comparing the activities at 20 MeV (Orsay), 50 MeV (Louvain-la-Neuve), 80 MeV and 130 MeV (KVI). The number of atoms of ^{90}Kr, ^{91}Kr, ^{92}Kr, ^{139}Xe, ^{140}Xe collected on the cold finger are reported in table 3.1. These values are not corrected for the decay losses due to the delays in the transfer line between the UCx- target and the cold finger. A gain in the production of noble gases by a factor of about 10 has been obtained between a measurement at deuteron energies of 20 MeV and 130 MeV. There is a fast increase up to 80 MeV which then slows down in the same way as the production of neutrons.

Energy MeV	^{89}Kr 3.15mn	^{90}Kr 32 s.	^{91}Kr 8.6 s	^{92}Kr 1.8 s	^{139}Xe 39.7 s	^{140}Xe 13.6 s	^{141}Xe 1.7 s
20		$3.0\ 10^5$	$8.3\ 10^4$	$1.6\ 10^4$	$2.6\ 10^5$	$7.2\ 10^4$	
50	$6.9\ 10^5$	$5.5\ 10^5$	$9.7\ 10^4$	$8.8\ 10^3$	$4.3\ 10^5$	$1.1\ 10^5$	$1.7\ 10^5$
80	$3.4\ 10^6$	$2.4\ 10^6$	$5.9\ 10^5$	$1.2\ 10^5$	$1.5\ 10^6$	$5.3\ 10^5$	$1.2\ 10^5$
130	$6.5\ 10^6$	$3.8\ 10^6$	$9.3\ 10^5$	$1.2\ 10^5$	$2.3\ 10^6$	$8.2\ 10^5$	$1.9\ 10^5$

Table 3.1 : Numbers of collected noble gas atoms, in atom per µC of deuteron beam for various energies. The converter was Be placed at 8 cm from the UCx target. Target temperature was 1800°C for all cases.

The influence of the geometry and the nature of the converter have been studied during the experiments at Louvain-La-Neuve and KVI at respectively 50 MeV and 80 MeV deuteron energy. The results obtained with a Be and a C converter placed at a distance of 8 cm from the UCx target are similar. A gain by a factor 3.2 has been obtained with a C-converter placed at a distance of 1.3 cm from the target. It is clear that, due to the finite width of the neutron angular distribution, the converter must be as close as possible to the target. Heat radiated by the target then becomes a problem for a Be-converter, owing to the low melting point of Be. The carbon converter is therefore the best.

The influence of the target temperature has been studied during the experiment at Louvain-La-Neuve, with a beam energy of 50 MeV. The UCx-target was successively maintained at temperatures of 1600 C, 1800 C and 2000 C. A gain in the production of noble gases by a factor of between 2 and 3 has been obtained between 1600 C and 2000 C.

3.2.2 Comparison with simulations

Simulations of this experiment have been performed at IPN-Orsay in order to determine the total efficiency of the process. Simulations have been made for a C-converter separated by 10 mm from an UCx-target of 7 mm radius and 60 mm length, the actual geometry of the target at PARRNe1. In this context, a new code FICNeR has been developed at IPN-Orsay (see annex 2). Comparison with data available at 80 and 130 MeV shows satisfactory agreement with respect to energy dependence. In these measurements at different energies the same scaling factor for release efficiency for a given element was used. There is a saturation of nuclide production at 100 MeV d-energy and even a decrease for the most neutron-rich isotopes. These results are easy to explain by combining the neutron production and cross section data, as mentioned in the previous section. Simulations of these experiments have been performed at GANIL with the LAHET + MCNP + CINDER codes, the in-target yields calculated with the codes are a factor 5-10 more than the measured ones indicating that the diffusion efficiency is about 10-20%. However, at 50MeV the in-target yields from simulation and out-target yields from experiments shows a good agreement. As the diffusion efficiency should be the same as at 80 and 130MeV experiments, this comparison seems to indicate that, at lower energy (50 MeV), the codes LAHET + MCNP + CINDER underestimate the in-target yields.

3.3 Production of ions in a prototype 1^+ ISOL system. Experiments with PARRNe2

The previous series of experiments concern only production and diffusion of noble gases Kr and Xe as atoms. They belong to the asymmetric fission. The next step is naturally to evaluate the production and diffusion not only of Kr and Xe, but of a wider range of nuclei with very different chemical properties. This can be achieved with an ISOL (Isotope Separator On-Line) system where fission products are delivered in the form of singly-charged ions. Until quite recently, no ISOL device has been available in Europe with deuteron energy in the range 20-200 MeV. However, the construction of the separator PARRNe2 at the 15 MV Tandem of Orsay has been completed in February 1999. This device is a prototype for the production of radioactive ions using fission induced by fast neutrons [LAU00]. It is also a fully equipped on-line mass separator permitting spectroscopic experiments. Deuteron energies up to 27 MeV and intensities up to 600 nA are currently available. Several experiments have been performed with different target configurations: a solid and porous high-temperature UCx-target and a molten uranium target. The first on-line test of the separator has been performed in April 99 with a UCx target with 33 g of uranium. It has been followed by others in May (250 g molten uranium target and December 2000 with an UCx target containing 70 g of uranium.

In the first on-line experiment with a UCx target, exotic isotopes of noble gases Kr (up to mass 94) and Xe (up to mass 141) beams were observed. The volatile elements Zn and Cd were observed with higher yields than expected, probably because the high energy tail of the neutron spectrum opens the symmetric and very asymmetric fission modes. Iodine is also separated. The alkalines Rb and Cs have lower yields than expected; however, they are quickly released. This is shown by the fact that the efficiencies deduced by comparison with the model are the same, 0.4%, for ^{95}Rb and ^{94}Rb although the radioactive half-life of the latter is only 0.4 s.

The December 2000 experiment was similar but with a heavier UCx target (70 g) and another ion source, the MK5 plasma ion-source designed at CERN-ISOLDE, instead of the Nier-Bernas discharge source originally used. All the elements Cu, Zn, Ga, Ge, As, Se, Br, Kr, Rb, Sr (light group) and Ag, Cd, In, Sn, Sb, Te, I, Xe, Ba (heavy group) were extracted as 1^+ ions with the fast release UCx-target. Compared with elements produced by ISOLDE at 1 GeV proton energy, only Ni and Pd (which are produced with very low cross section at the tandem since only the asymmetric fission mode is open at this low energy) and the Y, La, Ce, Pr, Nd, Pm and Sm group (which for diffusion needs to be in molecular form by adjunction of F or O) were not observed. With respect to the first experiment with a 33 g UCx-target, yields have improved by factors of about 20 for the elements already produced with high yields and up to 50 for others [IBR01]. Among the key results is the separation of the double magic ^{132}Sn produced, extracted and ionised in the 1^+ form with a rate of 3.5 10^5/s. Extrapolation to 100 MeV, 500 µAe allows us expect about 1.7 10^9 1^+ ions/s of ^{132}Sn.

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Annex 4

4 Choice of deuteron energy

This chapter deals with the best deuteron energy for the case where it is used on a converter. We first summarise the conclusions from previous sections. Neutron production increases with energy, at first rapidly, but more slowly beyond 100 MeV. The neutron energy distribution can be decomposed into a forward-peaked component of energy centred at 0.4 times the deuteron energy and a more isotropic one of a few MeV. The forward-peaked component is of interest since the target is located downstream of the converter. The total cross section for neutron-induced fission peaks at 40 MeV, i.e. a deuteron energy of 100 MeV. In addition, higher energy leads to increased probability for neutron evaporation before fission, which shifts the isotopic distributions closer to the valley of beta-stability. Finally, it has to be considered how the beam power could be dissipated. This issue is addressed in annex 5 where it is concluded that high energy and low intensity is to be preferred over high intensity and low energy. The result of these conflicting trends is the existence of a maximum production for certain energy and intensity combination.

The number of fissions as a function of deuteron energy has been calculated with the LAHET+MNCP and FICNeR codes at GANIL and Orsay. In the former, neutrons generated in the converter as well as those of lower energy produced in the target by evaporation and fission are considered as able to induce fission in the target. In the latter, only the neutrons generated in the converter are considered. Figure 4.1 shows that at same beam power:

- The number of fissions per absorbed beam power calculated by LAHET taking into account all the incident neutrons increases strongly up to 100 MeV, and thereafter the gain is rather negligible.

- However, if an upper cut-off of 20 MeV is applied for the neutron energy in order to select fission leading to the most exotic nuclei, the results with FICNER and LAHET are very comparable for roughly the same UCx-target length. There is then a decrease by roughly a factor 2 from 100 MeV to 200 MeV for both, (the geometry used for FICNeR is reported in reference [MIR00] and the one for LAHET in annex 5).

- The number of fissions per absorbed beam power calculated by LAHET for all the neutrons and if a cut off of 20 MeV is applied, differ by almost an order of magnitude above 100 MeV deuteron energy.

In order to get a constant number of total fissions per second (10^{13} fissions/s) and for a 13 g/cm^2 UCx-target, the primary beam intensity should be 2174, 714, 505, and 241 μA at 50 , 80, 100 and 200 MeV, respectively. Figure 4.2 shows the rates for some nuclides calculated with LAHET in these conditions. The curves mirror the influence of beam intensity, total cross section, opening of fission modes and shifts of the centres of A(Z) distributions. The light and heavy isotopes ^{78}Zn and ^{156}Sm, at the edges of the mass distribution, are produced only at the highest energies when the channel for super-asymmetric fission is open. The not very n-rich nucleus ^{112}Rh has a similar, although less dramatic, trend due to the opening of the symmetric channel. The pairs of isotopes ^{88}Kr-^{92}Kr and ^{138}Xe-^{142}Xe demonstrate the shifts of the distributions towards the valley of β-stability

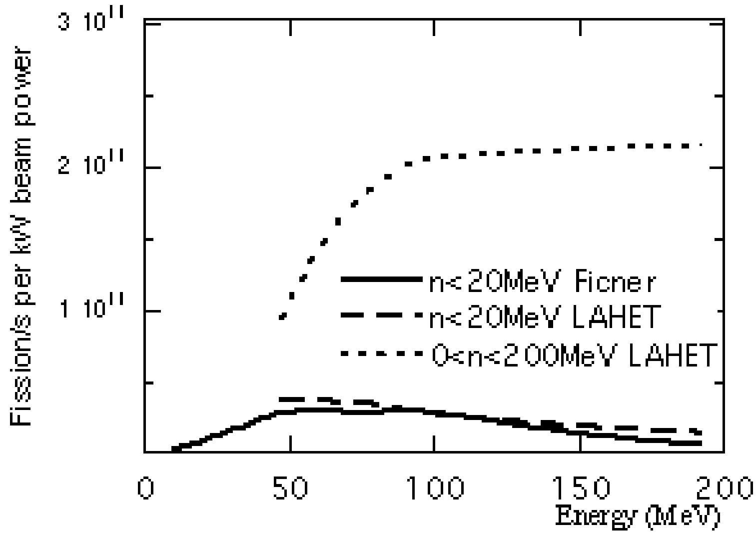

Figure 4.1: Calculated number of fissions per second and per dissipated beam power in the same target as function of deuteron beam energy. The calculation is for a deuteron beam intensity of 100 µAe and a target of about 13 g/cm^2.

With increasing energy, as the mass distributions widens and fills the valley in the symmetric region. There is no all-purpose deuteron energy: for the asymmetric region, a lower energy, e.g. 50 MeV, is suitable while for other fission modes higher energy is preferable. However, the prediction of LAHET is to be regarded with some caution for the super-asymmetric because this mode is probably not properly included in the code. In the symmetric-fission region, in-target rates are reasonable but most of the elements, being refractory, are almost impossible to get out of the target by the ISOL method.

This suggests that there is no need to go beyond 100 MeV and a somewhat lower energy is acceptable. With respect to beam power dissipation the range between 80 and 100 MeV is regarded as the one to be chosen. At very low energy (50 MeV), the high required beam power (about 100 kW for 10^{13} fissions/s) is a serious drawback for dissipation of energy in a relatively short converter (see annex 5). Moreover, this energy range (80-100MeV) is very suitable with the safety of operation of the radioactive beam facility since the amount of undesirable α-activities is there at its lowest (see annex 8).

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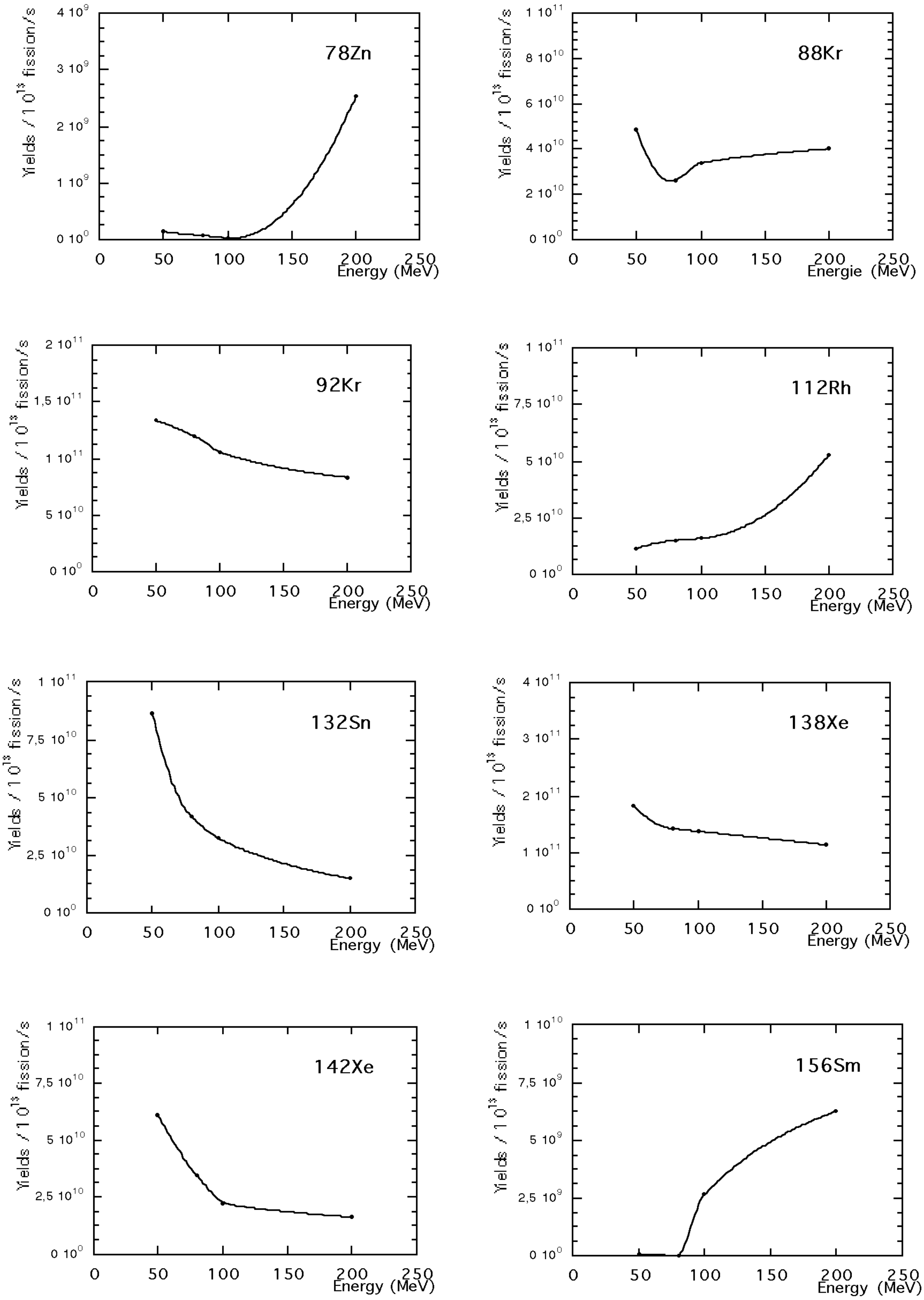

Fig 4.2: Yields for selected nuclei for a 10^{13} fissions rate.

Annex 5

5. Thermal properties and deuteron beam intensity

Here we describe the thermal properties of the converter and target assembly and estimate the maximum deuteron beam intensity required to reach the design goal of 10^{13} fissions / s.

5.1 Dissipation of the primary beam power

The highest primary beam intensity is desirable as it proportionally increases nuclide production. However, the energy deposited in the converter or the uranium target heats them and could damage the target. The incident beam power at which this becomes a real issue must be evaluated in order to assess the possible primary beam intensity. The goal is to study the temperature distribution inside the converter and/or target for different configurations. The maximum beam power that can be withstood by the target determines the achievable radioactive beam intensity.

The melting point of C is about 3500°C. It is thus high enough since a massive converter can be cooled. In contrast, those of target materials UC and UC_2 are only 2450°C and 2390°C, respectively. Since the target has to be maintained at a temperature at least of 2000-2200°C in order to favour fast diffusion but should remain below the material melting point, there is little room left for safe operation. Calculations have been made for three configurations, all with a UCx-target:

- the converter method: the deuteron beam is stopped in a C-converter. The radioactive elements are produced in the target by reactions induced by fast neutrons due to deuteron break-up only. The method has the merit of the lowest heat deposition depending on the beam (energy released by the stopping of the fission products) and the possibility to adjust the temperature with an external oven.

- the direct method: a target not thick enough to stop the deuteron beam. The beam is stopped in a beam catcher downstream. The target is heated mainly by the energy loss of the primary deuteron beam and, to a lesser extent, by fission. This configuration avoids the high density of energy deposited near the end of the beam range (the so-called Bragg peak).

- the direct method: a target thick enough to absorb all beam energy.

In the two last cases, a rather difficult compromise has to be found between primary beam intensity and target size in order to maintain the temperature in the acceptable range. Calculations were made at GANIL. First simple assumptions were made to get a rough estimate of heat transfer and temperatures. Then a more precise answer was obtained with the SYSTUS and Rubens codes for the most interesting configurations [NAY01], and also for the case where the Bragg peak is located inside the converter. All cases considered have a cylindrical symmetry axis along the beam direction. The emissivity of UCx compounds being unknown, the emissivity of C was used, which approximation can be justified as long as the proportion of uranium remains small (thus for x=9, there is 1 atom of uranium for 9 atoms of carbon).

5.1.1 Converter and target

These simulations have been carried out for a C-converter and d-beam of variable energy and different configurations. The main conclusions are the following:

- For same beam power (energy times intensity) the maximum temperature in the converter is the lowest at the highest energy. The energy of the highest-energy beam is dissipated over a larger depth while the converter length is increased in accordance with the deuteron range.

- The temperature at the centre of the converter decreases with increasing dimensions. However, the length cannot be exceedingly large. Too long a converter reduces the useful neutron flux by increasing the distance between the source of neutrons and the target.

- If the converter is cooled on the sides by circulating water. Then a conical converter with a larger outer surface than a cylinder is to be preferred owing to its lower surface temperature which is less stressing for water cooling. This type of converter allows higher beam intensity e.g. about 100, 200 and 400 µAe for energies of 50, 80 and 200 MeV, respectively, with the standard heat-exchange coefficient used at GANIL. Increasing the heat exchange coefficient also decreases the outside temperature. However, increasing outer surface or heat-exchange has little effect on the maximum temperature at the central region

- It is possible, by defocusing the beam and using a large radius, to achieve adequate temperatures of the surface for high beam power for a cylindrical converter too. At the same energies as above, intensities of about 50, 80 and 160 µAe are allowed with the standard heat exchange coefficient. By going a bit beyond the standard value it seems possible even to accept up to 400 µAe at 200 MeV. The limitation is indeed due to the cooling of the outer surface rather than the inner temperature which is still far from the melting point of graphite.

A detailed simulation has been carried out for a 100 MeV deuteron beam, this energy being near the best according to combination of neutron production and nuclide cross sections. A converter was designed with a special conical shape. The central part is the shortest, (3.5 cm) slightly longer than the range of deuterons (2.8 cm) to keep a wide solid angle for neutrons. The radius is 11 cm. The beam has a radius of 2 cm. The UCx fission target of 6 cm diameter is placed at 1 cm behind the converter. It is made of lamellae of 1 mm thickness, separated by 0.5 mm, distributed over a length of 8.5 cm and contains 360 g of uranium. It is surrounded by a container and an external oven. The whole is enclosed in an Al water-cooled box. The assembly is shown in Fig. 5.1. For this target size and the assumed beam intensities, the power due to fission is not enough to reach 2000^0C. It is therefore necessary to supply extra heat via the external oven. It is a C-cylinder separated from the target container by 0.5 cm with a surface temperature of 2200^0C. The converter temperature increases due to the vicinity of the hot target (by about 25^0C on the outer surface and 300^0C in the central part).
Finally, we estimate that it is possible to accept 350 µA of beam at 100 MeV. This gives 6.10^{12} fissions/s in this target. It is probably still possible to increase the beam power up to 50 KW (500 µAe) by faster circulation of cooling water and by a better adaptation of the mechanical supports of the converter. Yet, there remain an uncertainty related to the losses at the thermal contacts between the different parts. If a further defocusing of the primary beam is also possible, it implies a larger target size. This has the disadvantages of lowering the diffusion efficiency and being challenging to make. A simulation performed at IPN-Orsay suggests a beam of only 0.5 cm radius could be accepted by a C-converter up to 100 µAe. In this case, a UCx-target with comparable size to that used in PARRNe-2 experiments could be used. This way, yields could be determined without extrapolation concerning the diffusion process. The only obvious drawback is an overall lower production rate.

Further improvement in cooling could be achieved by using another refrigerant liquid, gaseous helium or a liquid converter. However, these latter two solutions are beyond the scope of R&D for a short-term facility.

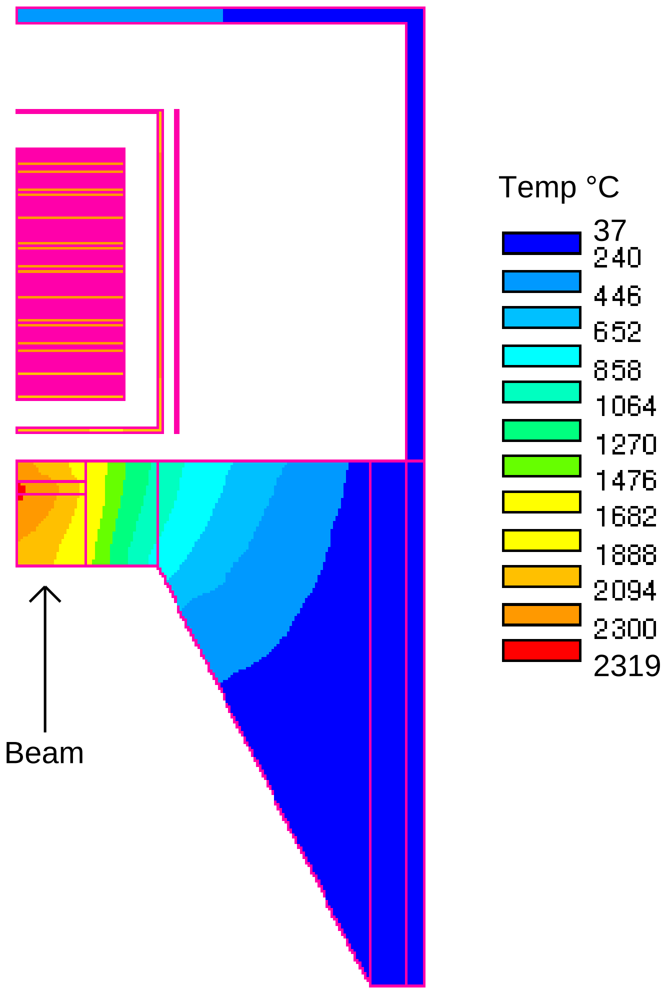

Fig 5.1 Conical graphite converter and uranium-carbide target assembly. The outer surface of the converter is water cooled with the standard heat exchange coefficient used at GANIL 0.5W/(cm^2 ^{0}C). The temperature distribution inside the converter has been calculated with the SYSTUS code for a 400 μAe deuteron beam of 100 MeV and defocusing to a radius of 2 cm (see text for more details).

5.1.2 Targets of various lengths without converter

In this case, a 100 MeV deuteron beam directly bombards the target. The power load in the UCx target is now much higher. Most of it is due to absorption of the deuteron beam. The heating due to fission is a small fraction. A working temperature in the 2000-2200^{0}C range is only obtained by defocusing the beam, sending it off the axis and rotating it in order to spread the thermal load. Influences of different parameters such as beam and target radius, beam rotation, conical or cylindrical target shape were simulated at GANIL. The main conclusions are the following:

- A temperature of 2000^0 C is already reached for 5 kW beam power for a beam radius of 2 cm as before but moved 1 cm off the axis and rotated. The target radius is thus 3 cm. This is possible only if the front end of the target is shaped as a cone of 20 degrees opening angle, extending the target length to 16.7 cm. The 50 µAe beam on this target produces 1.8 10^{13} fissions/s which cause an additional 0.6 kW power in the target.

- Under the same beam conditions, the cylindrical target used with the converter (see above) can take 3 kW beam power only. There are 1.0 10^{13} fissions/s which add 0.3 kW power in the target. The conical shape distributes the beam power better than the cylindrical one but unfortunately is more difficult to machine. If the target is not thick enough to stop the beam (3 cm), the deuteron beam maximum intensity is 50 µAe and produces 1.2 10^{13} fissions/s (0.4 kW additional power).

5.1.3 Outlook

For the deuteron beam of 100 MeV which seems to be the best choice for radioactive nuclide production, the maximum beam intensities are:

- 350 µAe with a C-converter. It also seems possible to reach 500 µAe, but cooling the outer surface of the converter has to be further investigated. Another solutions using an intermediate interface of another refrigerant should also be investigated. Another possibility is to use a rotating target like SISSI at GANIL. This is being investigated by the R3B RTD project.

- 50 µAe directly in the UCx target by allowing the beam to pass through. This configuration avoids overheating of the target that would have taken place near the end of the trajectory of the ions due to the Bragg peak. The large amount of deposited energy when the beam comes to rest is not useful for nuclide production.

- 30 µAe directly in the UCx-target which stops the beam.

The indirect method is the most reliable concerning the control of target temperature. The beam energy losses take place in the converter with a high melting point and not in the UCx target. This offers the possibility to adjust the target temperature with an oven independently of the beam power.

<u>5.2 Estimated yields for the three configurations</u>

The maximum beam intensities acceptable by the system were subsequently used for yield calculations performed by the GANIL group with the LAHET+MCNP+CINDER code for a deuteron beam energy of 100 MeV [SAI01]. The beam intensity and target size were chosen according to the temperature simulations in order to get about 10^{13} fissions/s. As above, targets are made of lamellae of 1 mm thickness. The composition is 1 atom of U for 9 atoms of C. The density of ^{238}U is 2.3 g/cm^3 as in the first PARRNe1 experiment. However, the target radius of 3 cm is larger than for the PARRNe1 target (0.8 cm) and the lamellae are more widely spaced (0.5 mm) than in the PARRNe target (0.1 mm) in order to enhance diffusion. The beam is defocused to 2 cm radius.

Three configurations were considered:

- Case 1: A carbon converter ahead of a UCx target with 56 slices. Their number of slices is comparable to the 50 slices in PARRNe1 experiments.

- Case 2: A UCx target with 20 slices, not thick enough to stop the beam, followed by a beam catcher.

- Case 3: The same target as in the first case in which the beam is stopped.

The "in-target" yields are compared in table 5.1. The main conclusions are the following:

- In case 1, a large amount of tritium is produced in the converter. In case 2, large quantities of α-emitters are produced in the target.

- Even through a somewhat lower number than 10^{13} fissions/s is obtained with 350 µAe in case 1, the nuclei are more neutron-rich than in case 2. This more than compensates for the smaller number of fissions. This is a consequence of the capture of a neutron instead of a deuteron before fission and of the lower projectile energy. Case 2 is better for the less exotic nuclides and those in the region of symmetric fission (Z=45). However, most of the elements in the symmetric region are refractory and there is little hope of extracting intense beams of them.

- The ^{132}Sn in-target production with 350 µAe (case 1) is 2.2 10^{10} /s. This should be compared with the rate of 1.7 10^{9} ions/s diffused and ionised which is extrapolated from the PARRNe2 measurements after scaling with the beam intensity and the estimated gain when increasing the beam energy.

- Case 3 is not very interesting due to the limited beam intensity and, more importantly, to the poor reliability of operation due to the narrow temperature range allowed.

<u>References</u>

[NAY01] D. Nayak, M.G. Saint-Laurent, F, Pellemoine and V. Dubois, rapport interne GANIL, March 2001

[SAI01] M.G. Saint Laurent et C. Stodel., rapport interne GANIL, April 2001

100MeV U/C =1/9 ρ(U)=2.3 g/cm^3 Fissions/s	d+C+UC 350 µAe 6.3 10^{12}	D+UC +C 50 µAe 1.2 10^{13}	d+UC 30 µAe 1.0 10^{13}
Total activity (Bq) 1 month after end of irradiation.	1.3 10^{12}	3.2 10^{11}	2.2 10^{11}
Dose rate (µSv/H) at 40 cm — Max 1 year after beam stop.	9.1 10^{6} 4.6 10^{2}	3.3 10^{5} 5.4 10^{2}	2.2 10^{6} 6.0 10^{2}
Tritium Max Bq	1.3 10^{10}	9.6 10^{8}	5.7 10^{8}
Alpha activity Bq 1 year after beam stop.	1.8 10^{7}	6.7 10^{8}	4.2 10^{8}
^{78}Zn	1.9 10^{9}	1.8 10^{9}	1.3 10^{9}
^{88}Kr	2.3 10^{10}	4.1 10^{10}	2.8 10^{10}
^{92}Kr	7.3 10^{10}	7.8 10^{10}	6.0 10^{10}
^{112}Rh	1.1 10^{10}	3.7 10^{10}	2.5 10^{10}
^{132}Sn	2.2 10^{10}	5.5 10^{9}	6.6 10^{9}
^{138}Xe	9.5 10^{10}	1.2 10^{11}	8.7 10^{11}
^{142}Xe	1.5 10^{10}	9.1 10^{9}	7.8 10^{9}
^{150}Ce	2.3 10^{10}	1.8 10^{9}	1.4 10^{10}
^{156}Sm	1.8 10^{9}	2.9 10^{9}	1.8 10^{9}
^{158}Sm	5.6 10^{9}	3.6 10^{9}	2.5 10^{9}

Table 5.1: In target-yields/s for selected nuclides calculated with the LAHET+MCNP+CINDER codes, taking into account the constraints due to thermal properties.

Annex 6

6. Charge breeding and post-acceleration

Ion sources used for mass separation deliver singly-charged ions. However, high charge states are required for injection into a cyclotron post-accelerator in order to reach the final energy (proportional to the square of the charge state for a cyclotron). Therefore charge breeding is necessary. In order to allow a variety of experiments, the energy of the radioactive beam must exceed the Coulomb barriers. The possibilities for post acceleration to suitable energies have been investigated.

6.1 1^+ / N^+ charge breeding

In the hostile highly radioactive environment near the target, it is logical that only a rather inexpensive 1^+ ion source can be installed. Charge breeding requires an expensive source that cannot routinely be exchanged. It should therefore operate in a shielded location. The resulting efficiency for N^+ ions is the product of the two efficiencies, for 1^+ ionisation of the elements diffused out of the target and for $1^+/N^+$ charge breeding. This shows that high efficiency is both processes is very important.

6.1.1 Ion-sources for singly-charged ions

There are no universal 1^+ ion sources with high efficiency for all elements. Different techniques have to be used depending on the chemical properties and ionisation potentials. Presently, many elements can be delivered by mass separators based on the ISOL method. At least 4 types of ion sources appear to be necessary for this purpose:

- An ECR ion source like MONO1000 for gaseous elements and some condensable elements (recent off-line tests gives close to 100% for singly-charged Ar and 41% for S);
- A hot plasma ion source like MK5 designed at ISOLDE with a hot transfer tube for condensable elements (40% efficiency for Sn), routinely used at ISOLDE and very recently at PARRNe-2;
- A surface-ionisation source for alkalines (near 90% efficiency for Rb and Cs);
- A laser source when metallic beams with high purity are required (2.5% efficiency for Sn).

6.1.2 Charge breeding

The efficiency for $1^+/N^+$ charge breeding is still low nowadays. An estimate can be deduced from the data measured by Chauvin et al. [CHA00] for different elements. Improvements of the breeding efficiency are foreseeable with the Phoenix source. Table 6.1 shows that Kr and Ag are obtained in higher charge states with higher efficiency than with the Minimafios source.

Nucleus	q	efficiency	Ion source
^{64}Zn	11+	3.0 %	DANFYSIK + Minimafios
^{84}Kr	11+	9.0 %	MONO1000 + Minimafios
^{84}Kr	14+	10.3 %	Phoenix Booster
^{85}Rb	15+	5.5 %	surface ionisation + Minimafios
^{107}Ag	17+	3.2%	microgan + Minimafios
^{107}Ag	19+	3.8 %	Phoenix Booster
^{134}Xe	16+	6.2 %	MONO1000 + Minimafios

Table 6.1. Efficiencies for charge breeding for some elements and various sources [CHA00].

6.2 Post acceleration

Depending on the energy requested for experiments, the radioactive beam at GANIL could be accelerated in the new cyclotron CIME or a combination of accelerators CIME + CSS2 or even C0 injector and CSS1 + CSS2. Due to the inherent problems of operating several cyclotrons in series with very weak beams, the latter solution appears to be difficult. Alternatively, a new machine could be an interesting solution. These possibilities were investigated at GANIL [BAR01].

6.2.1 With CIME

The CIME compact cyclotron has been designed for acceleration of ions for SPIRAL-1. With today's standard sources for multi-charged ions this restricts its use to ions of mass not very much higher than 100. The energy per nucleon in a cyclotron being proportional to $(q/A)^2$ it is necessary to increase the ionic charge state q with the highest probability in the distribution. Higher charge states may soon be available with high efficiency with new sources, e.g. the above mentioned Phoenix or SERSE at LNS-Catania. However, one must assume that 10 MeV/A is to remain a limit for some time. Table 6.1 shows for some nuclei of interest the energies which can be obtained with the high charge states of modern N^+ sources.

6.2.2 Coupling CIME and CSS2

Independent operation of both cyclotrons by debunching the beam at the exit of CIME and rebunching before CSS2 turns out to be technically impossible. It is, however, possible to run both machines at the same frequency, just running on different harmonics. This can work, provided the beam out of CIME is stripped to increase its charge state, say by a factor of 2. Stripping is plagued by a loss of 80% of the beam, which could be critical in case of weak radioactive beams extracted from the ion sources. The operating range of the combined machines would be from 11 to 22 MeV/A. This is enough to surmount the Coulomb barrier on all elements for all nuclei produced by fission and to perform various types of reactions with neutron-rich beams.

Element	charge	E/A (MeV/A)	VCb (MeV/A) on C	VCb (MeV/A) on Pb
^{68}Ni	13	9.6	2.9	5.0
^{78}Ni	13	7.3	2.7	4.4
^{90}Kr	15	7.3	3.3	5.0
^{94}Kr	15	6.7	3.3	4.8
^{128}Sn	18	5.2	4.1	5.2
^{132}Sn	18	4.9	4.1	5.1
^{140}Xe	20	5.4	4.4	5.3
^{144}Xe	20	5.	4.3	5.2

Table 6.2 Energies per nucleon delivered by CIME for realistic charge states in a near future. Coulomb barriers on carbon and lead in MeV/nucleon are also shown.

6.2.3 Outlook

The GANIL group has also investigated the problems related with the transport of a 1^+ ion beam to CIME and the site for the SPIRAL-II cave. A new cave is definitely the favoured option.
The energies reached by CIME alone will remain below 10 MeV/A while the CIME + CSS2 combination will allow the 11 – 22 MeV/A range. There will be therefore a gap. Higher energies can in principle be reached combining the 3 GANIL cyclotrons, but this is evaluated as being very difficult indeed, owing to the lack of beam diagnostics suitable for the very low intensity beams.

References

[CHA00] N. Chauvin, PhD ISN Grenoble, ISN00-76 (2000)
[BAR01] E. Baron, rapport interne , February 2001 and E. Baron, report to the SPIRAL phase II steering committee, April 2001

Annex 7

7. The deuteron driver for the SPIRAL-II project

7.1 Beam energy and intensity

In the choice of the deuteron energy a number of conflicting aspects have to be considered. From arguments mentioned previously, it has been concluded that the optimum energy is around 80-100 MeV. The goal of 10^{13} fissions/s can be reached with a deuteron beam intensity of 0.5 mA.This is a realistic beam intensity for a cyclotron. A linear accelerator could even provide significantly higher currents.

7.2 Accelerator type

From a technical point of view both a cyclotron and a linear accelerator are suitable to produce the deuteron beam needed. For cyclotrons the presently achieved maximum beam intensities are about 1 mA of 30 MeV protons in commercial compact cyclotrons used for isotope production and 2 mA of 72 (and 590) MeV protons in the separated-sector cyclotrons at PSI (Switzerland).
For linear accelerators the achievable currents are significantly higher. However, at the low energy (about 80 MeV) needed for the present application and assuming that the required intensity can be provided by a cyclotron, linear accelerators are much less cost-effective. They have a larger footprint, thus requiring a higher investment for infrastructure (building etc.), while the accelerator itself is also far more expensive than the equivalent cyclotron. Finally, if continuous-wave operation is required only a linear accelerator with superconducting radio-frequency cavities can be used, which further increases the investment.
It is concluded that for a short-term project, the use of a cyclotron is the best option: it permits us to achieve the objectives of the project at the lowest costs. However, as a consequence of this choice, any significant increase in intensity in terms of a future upgrade will require large additional investments.

7.3 Cyclotron characteristics

Radiation safety is a very important issue when accelerating deuterons to an energy of 80-100 MeV with an intensity up to 0.5 mA. Beam losses around 1% already cause strong activation of the parts of the cyclotron on which the lost beam impinges. Furthermore a large flux of high energy neutrons is produced, which will significantly activate the cyclotron and its surroundings. This flux of neutrons is increased even further because of the low threshold for neutron production in the $d(d,n)^3He$ reaction on lost deuterons that have been implanted in various parts of the cyclotron. Furthermore tritium is produced via the reaction $d(d,p)^3H$. This may lead to additional radiation safety problems.
It is thus essential that a very high transmission ($\geq$ 99.9%) is achieved, in particular for the later stage of acceleration and for the extraction. It is the prime factor in assessing the suitability of the three possible systems studied:

- the GANIL cyclotrons
- the SARA booster-cyclotron with a new injector

- a new cyclotron

7.3.1 The present GANIL cyclotrons

The GANIL cyclotrons can accelerate deuterons to energies in the range 12 to 26 MeV and around 80 MeV. With the injector cyclotron and the first separated sector cyclotron the energy range 12 to 26 MeV can be covered without modifications of the accelerators. Let us remember that the fission yield at 26 MeV is at least an order of magnitude lower than at 80 MeV, taking into consideration the energy dependence of the neutron production and fission cross sections.
Around 80 MeV acceleration of deuterons using the complete chain of injector cyclotron and two separated-sector cyclotrons is possible. In the injector cyclotron and the first separated-sector cyclotron molecular d_2 ions would be accelerated, and would subsequently be stripped into two deuterons on injection into the second separated sector cyclotron. Acceleration has been tested at the rather low magnetic field required using ^{16}O. This test has shown that acceleration is possible but losses in the injector are too large. As the neutron production by deuterons at the energy of 80 MeV is higher than with the usually accelerated ions at GANIL, radiation safety problems will impose much more serious constraints, connected to both the shielding and the activation of components.

7.3.2 The SARA booster-cyclotron with a new injector

The SARA booster-cyclotron is a separated-sector cyclotron, which is presently being decommissioned at the Institut des Sciences Nucléaires in Grenoble (France). It can accelerate deuterons to a maximum energy of 72 MeV, slightly lower than the value aimed at here. It has an energy gain of a factor 5, so that an injector (cyclotron) delivering 14.5 MeV deuterons is needed. The matching conditions between the injector and booster cyclotron strongly constrain the characteristics of the injector. Consequently the injector would have to be specifically developed. Furthermore the injection, RF-system and extraction of the booster would have to be reconstructed to meet the requirements of the high intensity operation.
The requirements on the extraction efficiency can only be met by the use of stripping extraction of negative ions. These negative ions thus have to be extracted from the injector by a standard extraction system using an electrostatic deflector. Taking into consideration the lower yield and energy of neutrons produced at 14.5 MeV as compared to 80 MeV the extraction efficiency of the injector should be at least 95%, which is close to the limit of feasibility.

7.3.3 A new cyclotron

An analysis of the existing cyclotrons delivering high intensity proton beams (≥ 1 mA) at a fixed energy shows two possible schemes to achieve the required transmission:

- A large, low-field separated-sector cyclotron with high energy gain per turn and 'classical' extraction (*e.g.* PSI injector II: 2 mA, 72 MeV protons). Cyclotrons of this type have been developed exclusively for research institutions. The high transmission is obtained by maximising the radial distance between subsequent turns in the machine, so that an electrostatic septum, which bends away the last turn, can be inserted in between two turns. A cyclotron for 80 MeV deuterons based on this approach would have an extraction radius of 3.5 m.

- A compact cyclotron accelerating negative ions with stripping extraction (*e.g.* CYCLON30 from IBA (Belgium) and TR30 from EBBCO (Canada); both delivering 1 mA 30 MeV protons). Around 20 cyclotrons of this type are used routinely for isotope production in an industrial environment. The efficiency of the extraction process is essentially 100%; the overall transmission is determined by beam losses during acceleration caused by the magnetic field and the interaction with the residual gas.

A cyclotron based on these examples would have an extraction radius of about 1.6 m. For the present application the superior beam quality of the first, far more expensive scheme is not required. Furthermore, the use of stripping extraction makes it possible to vary the energy over a range of roughly a factor two (40 – 80 MeV) by changing the radius at which the stripper foil is located.

7.4 Analysis and recommendation

The use of the present GANIL-cyclotrons requires by far the lowest investment. The investment for a dedicated accelerator, a new compact cyclotron or the SARA booster + a new injector, is estimated to be of the same order of magnitude (10 – 12 MEuro without infrastructure costs). The installation of a dedicated accelerator will make the operation of the SPIRAL-facility (and of the GANIL-facility in general) more flexible, efficient and versatile:

- Development work on beams with SPIRAL-II can proceed while beams from SPIRAL-I or the present GANIL-facility are delivered for experiments.

- Operation of the SPIRAL-facility is decoupled from that of the present GANIL-cyclotrons.

- The intensity of the radioactive beams attainable is at least an order of magnitude higher, thus significantly extending the range of feasible experiments.

- The possibility to inject beams from SPIRAL-II into the present GANIL-cyclotrons remains open.

- Simplicity of both concept and operation is an important asset for the deuteron accelerator in the SPIRAL-II project. The compact cyclotron accelerating negative ions has by far the best score on this aspect.

- The development and construction connected to the use of the present GANIL-cyclotrons or the SARA-booster will require a large effort from the accelerator staff of GANIL (or other possible partners in the project). The development and construction of the compact cyclotron, based on existing industrial designs, is suitable for contracting to industry.

It is thus concluded that a dedicated compact cyclotron accelerating negative deuteron ions is the optimal solution for a short-term project.

Annex 8

8. Safety aspects

Today there does not exist an accelerator with such high primary beam intensity (500 µAe), able to deliver deuterons with energies from a few MeV up to 200 MeV. With respect to safety consideration two issues were investigated by the GANIL group :

- Simulation of activity as a function of energy for different configurations of converter and target and deuteron energies.

- Experiments in order to determine the attenuation length of neutrons in concrete, the air activation, the development of a method to measure the tritium diffused out of the converter or target.

Moreover, the Orsay group at the rather low incident energy available at the Orsay tandem has performed measurements of α emitters produced in the target, and residual activities in the oil of the vacuum pumping system.

8.1 Simulations of activities due to critical radionuclides

Comparison of nuclide production yields and activities for various converters and energies were performed at GANIL with the LAHET-MCNP-CINDER code. Even if this code underestimates globally the production yields, comparison of relative values allows us to determine advantages and disadvantages of different configurations. Special software has been developed in order to facilitate the evaluation of the simulations with the LAHET code [CAS00]. One must consider the total activity, the radiation doses due to β and γ-activities as well as the long–lived tritium and α-emitters produced by reactions in the converter and target. Moreover, special attention has to be devoted to those nuclides whose rate could be above the Euratom norms. The calculation have been constrained by the efficient production of a number of isotopes of rare gases Kr and Xe in the asymmetric fission region, the doubly magic ^{132}Sn, ^{78}Zn in the super-asymmetric region and ^{112}Rh in the symmetric region. The simulations have been carried out for deuteron energies in the 50 to 200 MeV range for Li, Be and C converters and a UCx converter-target. It has to be noted that the Monte Carlo method used in the MCNP code requires a large amount of events to smooth out statistical fluctuations. It is therefore more appropriate to discuss general trends rather than elaborate on local strong variation for a weakly produced nuclide:

- It is clear that α-activities are produced in large amounts mainly at the highest energies since reaction channels for actinides are not open at low energy. This implies serious constraints concerning the confinement and a risk analysis should be done for hypothetical accidents. The α-activity is higher for deuteron beam on a UCx target in the direct method than when a converter is used. In the latter case, reactions in the fission target are due to neutrons only. They do not produce nuclei of higher Z than uranium, except via β-decay of uranium isotopes.

- Tritium production is larger with light-Z converters (C, Be or Li) than without. Carbon is the lowest producer of tritium among them.

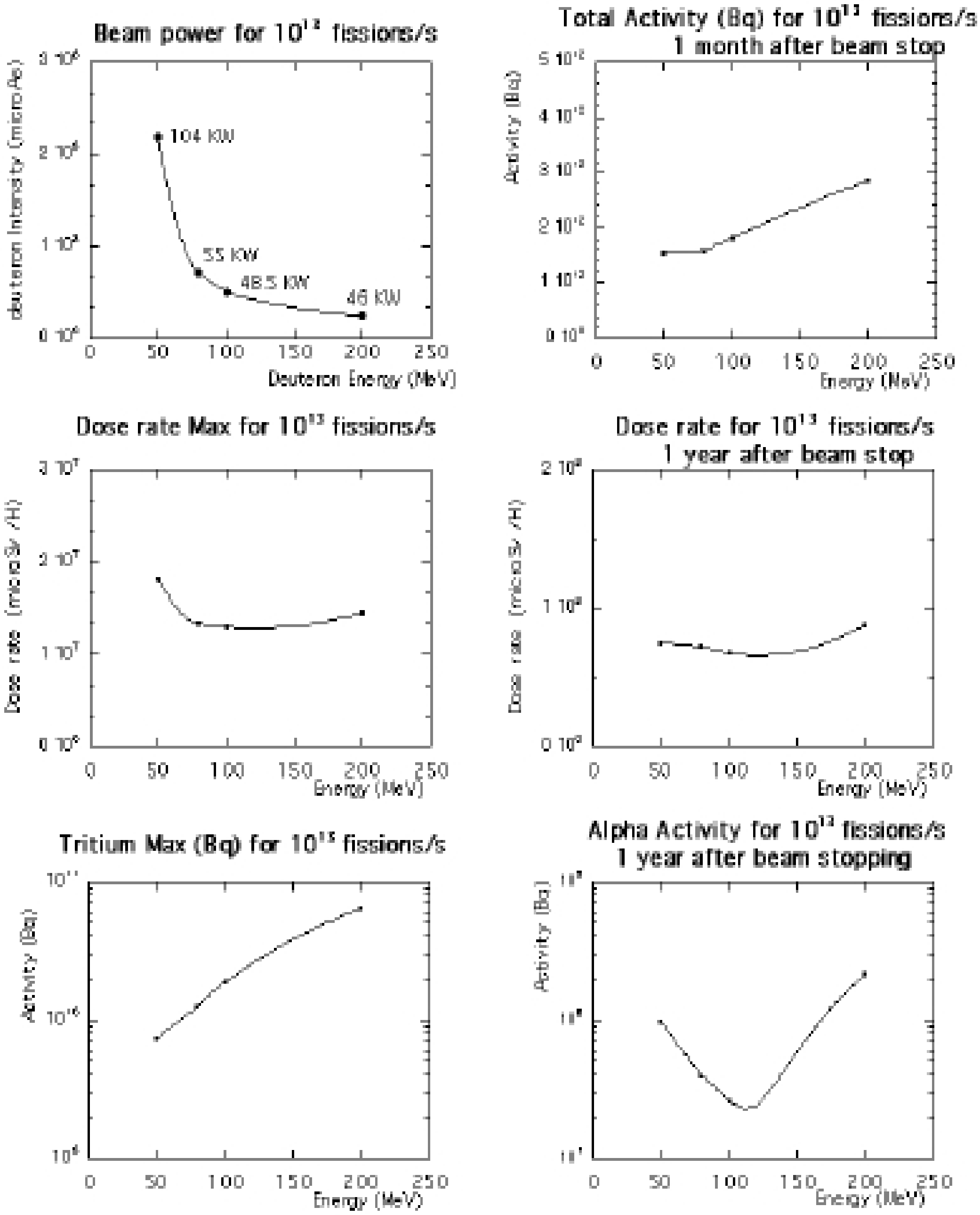

Fig. 8.1: Top, left: Beam intensity and power versus energy to obtain 10^{13} fission/s . Other pictures show, for this number of fissions, the radiation doses at 40cm during operation of the facility, the rate of tritium and of long-lived activities after shut down.

A goal of at least 10^{13} fissions/s has been fixed by the International Advisory Committee of SPIRAL-II. Fig. 8.1 displays the trends of selected set of important parameters for the production
of critical activities. These curves are normalised in order to kept the number of fissions per second constant. The top left figure shows that high power and high deuteron-beam current are needed at 50 MeV energy. The values at 100 and 200 MeV are rather similar. They require only half the power and about one fourth of the beam intensity needed at 50 MeV. This does not favour the choice of 50 MeV as a design goal for a cyclotron as many other parameters are only rather weakly dependent on the energy between 50 and 100 MeV. Only the production of tritium and the total activity one month after the end of irradiation are increasing with energy in that range. Even more remarkably, the dose rate during operation of the facility and the residual α-activity 1 year after beam cut-off have a minimum near 100 MeV. This feature is due to the opposing trends of decreasing input power and increasing cross sections with energy. It is therefore without any doubt that a deuteron energy near 100 MeV is the best one with respect to the minimisation of production of undesirable activities. Production of critical activities are reported in table 8.1 for a 100MeV-500μA deuteron facility onto a Carbon –UCx-geometry as described in annex 5 §5.1.1.

	Converter	Target
Nuclei number above Euratom norm	13	
Maximum total Activity (Bq)	$8.1\ 10^{13}$	$3.3\ 10^{13}$
Total activity 1 year after beam cut-off (Bq)	$3.7\ 10^{10}$	$1.1\ 10^{10}$
Maximum tritium activity (Bq)	$1.9\ 10^{10}$	$2.2\ 10^{8}$
Maximum β−γ activity (Bq)	$6.4\ 10^{13}$	$3.2\ 10^{13}$
Maximum γ dose rate (μSv/h)	$9.9\ 10^{6}$	$3.4\ 10^{6}$
γ dose rate 1 year after beam cut-off (μSv/h)	$1.3\ 10^{2}$	$5.4\ 10^{2}$
Maximum α activity (Bq)	-	$1.1\ 10^{12}$
Maximum ^{235}U activity (Bq)	-	$6.7\ 10^{10}$
Maximum ^{237}U activity (Bq)	-	$9.6\ 10^{11}$
α activity 1 year after beam cut-off (Bq)	-	$2.7\ 10^{7}$
^{238}U activity (Bq)	-	$2.3\ 10^{7}$

Table 8.1 : Production of critical activities are reported in table 1 for a 100MeV-500μA deuteron facility onto a Carbon –UCx-geometry as described in annex 5 §5.1.1

8.2 Experiments

An important point related to the safety and the cost of an installation is the protection by concrete shielding, the ventilation of air near the production site and the activities trapped in the vacuum pumping system.

8.2.1 Attenuation length of neutrons in concrete

Angular distributions of neutrons have been measured and simulated for different converters. Most high-energy neutrons are emitted in a narrow forward cone in the direction of the target. In addition, there are lower energy neutrons (of a few MeV) due to neutron evaporation and fission in the converter and target that are emitted rather isotropically. Shielding is thus required not only at forward but also at backward angles. In order to correctly dimension the installation, the attenuation length of neutrons has been measured at GANIL for 'hematite' concrete, which is more effective than conventional concrete. Neutrons were produced by the reaction (75 MeV/A) ^{13}C + Cu with a beam power of 2 kW. Five bismuth and cobalt activation-detector pellets were placed inside concrete blocks at intervals of 20 cm along the radial direction [MAN99]. After irradiation, the quantitative analysis of isotopic activities produced by activation of Bi and Co yields neutron flux parameters, i.e. intensity and the shape of energy spectra. The latter is possible since reaction channels have specific thresholds and the energy dependence of cross sections is known. Data at various depths in the concrete block define the attenuation length. The angular dependence is also studied. Similar measurements were done previously at SATURNE (Saclay) with 100 MeV d on Be and U and 95 MeV/A ^{36}Ar on C [PAU98]. The concrete attenuation length determined in the experiment at GANIL are shorter by 15% than at SATURNE. This might be due the different arrangements of the concrete blocks, the shielding of detectors from scattered neutrons being less effective at SATURNE. Attenuation lengths are about 20 cm. They increase with the distance from the source and with energy. Typically, 80 cm of concrete are needed to attenuate the flux by an order of magnitude. These measurements give confidence in the estimates of concrete shielding to be used.

8.2.2 Evaluation of tritium production

Tritium can diffuse easily at temperatures above 400 degrees. This diffusion induces contamination of various elements (i.e. beam line, pumping system, etc.) and generates radioactive waste. Owing to its radioactive properties (a β-emitter with only 18 keV end-point energy), tritium is very difficult to detect. Consequently, its management is very constrained, even more than its radiobiological effects.
The LAHET code has been used for calculating the production of ^{3}H in the converter and target (see annex5 table1). However, of concern is only the amount which escapes from the converter or target by diffusion. Therefore, a method was designed at GANIL to measure the amount diffusing out of the converter-target [MAN99, STO01]. The principle of measurement is to collect the tritium on a cold-finger, using activated carbon. After release, the gas is mixed in a proportional gas detector to count the beta-particles emitted by its decay. A measurement has been performed at GANIL in order to test this method. A 95 MeV/A ^{24}Mg beam with 285 W power was sent into a hot and porous C target particularly favourable to diffusion. The collected amount of ^{3}H is 27% of the estimated total activity produced by the target fragmentation at the target temperature of 1350°C. By scaling to the production of 100 MeV d on C with LAHET code and assuming a release of 30%, the Euratom exemption threshold of 1GBq, is reached within 10 days of beam irradiation with 90 µA. This emphasises the need for a non-porous and cooled C-converter together with a special authorisation of radioactive release.

8.2.3 Activation of air in the experimental cave

Activation of air in the experimental cave produces light radioactive isotopes as ^{14}O, ^{15}O, ^{13}N, ^{11}C with relatively short lifetimes. Consequently, a radioactive release in the atmosphere could be possible if this happens after a reasonably delay to be determined after the beam shut down. However, depending on the quantities, radioactive release authorisation could be necessary.
Activation of air in the experimental cave has been measured at Louvain-La-Neuve for 50 MeV/A deuterons in a Be target by Dhilly et al. [DHI00]. The air in the vicinity of the target was extracted during irradiation and the activity of the flowing air was counted. Two ionisation chambers were used, one for the activated air, the other for reference of background. After beam shut-down the decay curves were analysed to infer the composition of the radioactive isotopes

(^{14}O, ^{15}O, ^{13}N, ^{11}C) during irradiation, owing to their different radioactive half-lives. The results (see table 8.2) allow us to check and validate the safety code for air activation with deuteron beams. The experimental measurements for SPIRAL (^{36}Ar (95 MeV/A) + C) with projectile fragmentation are also reported for comparison. An enhancement of a factor 3 is observed between SPIRAl-I and SPIRAL-II. This implies that a public inquiry and a radioactive gas release authorisation are necessary for a facility using 100 MeV, 500 μA deuteron beam.

reaction	^{14}O $T_{1/2}$=1.2 min	^{15}O $T_{1/2}$=2.1 min	^{13}N $T_{1/2}$= 10 min	^{11}C $T_{1/2}$= 20 min	global dose rate μSv.h-1/μAe
^{36}Ar (95 MeV/A) + C	10%	30%	35%	25%	3
d (25MeV/A) + Be	60%	29%	11%	0%	10

Table 8.2: Measured air activation (isotopic proportion and global dose rate per incident μA) for d (25 MeV/A) and comparison with SPIRAL (^{36}Ar (95 MeV/A) + C).

8-2-4 Evaluation of concrete protection

Concrete protections have been evaluated for the target-ion source cave assuming a neutron dose rate of less than 7.5 micro Sv/h outside the cave. The differential neutron distribution (in angle and energy) were calculated with the LAHET code. Calculations were performed for a 2mA deuteron beam as a function of the variable deuteron energy at 80 MeV, 160 MeV and 200 MeV. About 5m and 7m of concrete (density= 3.4 g.cm^{-3}) are required at forward angles for a deuteron beam, of respectively 80MeV and 200MeV, stopped in thick C-converter. Only 2 and 3m are needed at backward angles.

Calculations were also performed for a 2 mA deuteron beam with an energy of 80 MeV stopped in a thick Copper Faraday cup at the cyclotron centre and in the deflector at the cyclotron output. About 4.4m and 4.65m of concrete (density= 3.4g.cm^{-3}) are, respectively, required in order to have a neutron dose rate less than 2.5 microSv/h outside the cyclotron cave.

The estimate price is in the range of 3.8 Meuros.

8-3 In summary

Increasing the deuteron energy increases the overall production, and in particular that of neutron-rich nuclei in the symmetric region. But higher energy opens new channels for production of long-lived α-emitters which cause a problem of contamination in the long term. From these considerations it is recommended that one should use an energy of about 100 MeV or a bit less. This is also the energy desirable from the point of view of production of radioactive isotopes of interest. For a facility using a 100 MeV, 500 μA deuteron beam, a public inquiry and an authorisation for radioactive gas release are necessary. The α-emitter production implies serious constraints.

References

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Annex 9

9 In-target yields expected with a 100 MeV-500 μA deuteron facility

The in-target yields expected are calculated with the LAHET code in the case of the converter geometry described in annex 5 for a 100 MeV-500 μA deuteron facility. For each nucleus, the expected cumulative yields, parent production and the direct production are reported in table 9.1. The production after diffusion, ionisation and acceleration are reported in annex 10 for some elements.

Nuclei	Cumulative production	Parents Production	Direct production	Nuclei	Cumulative production	Parents Production	Direct production
Ni67	3,08E+05	2,80E+05	2,80E+04	**Ru105**	1,99E+11	1,99E+11	0,00E+00
Ni68	4,01E+05	2,94E+05	1,07E+05	**Ru107**	1,54E+11	1,30E+11	2,36E+10
Ni69	5,00E+05	2,49E+05	2,50E+05	**Ru108**	1,14E+11	7,17E+10	4,21E+10
Ni70	5,66E+05	1,46E+05	4,20E+05	**Ru109**	7,99E+10	6,14E+10	1,85E+10
Ni71	5,07E+05	6,34E+04	4,44E+05	**Ru110**	5,52E+10	2,56E+10	2,95E+10
Ni72	4,06E+05	1,85E+04	3,88E+05	**Ru111**	5,54E+10	1,40E+10	4,14E+10
Ni73	6,21E+05	1,13E+04	6,10E+05	**Ru112**	5,35E+10	3,68E+08	5,31E+10
Ni74	2,75E+05	1,54E+03	2,73E+05	**Ru113**	1,37E+10	2,74E+09	1,09E+10
				Ru114	2,90E+10	2,65E+09	2,64E+10
Cu71	7,06E+05	5,07E+05	1,98E+05	**Ru115**	2,77E+09	2,42E+06	2,77E+09
Cu72	8,19E+05	4,06E+05	4,13E+05				
Cu73	2,41E+06	6,21E+05	1,78E+06	**Rh107**	1,57E+11	1,54E+11	2,63E+09
Cu74	2,23E+06	2,75E+05	1,96E+06	**Rh109**	8,78E+10	7,99E+10	7,88E+09
Cu75	2,63E+09	9,58E+04	2,63E+09	**Rh111**	9,74E+10	5,54E+10	4,20E+10
Cu76	2,63E+09	2,53E+04	2,63E+09	**Rh112**	6,93E+10	5,35E+10	1,58E+10
Cu77	1,76E+06	9,64E+03	1,75E+06	**Rh113**	7,68E+10	1,37E+10	6,32E+10
Cu78	1,15E+06	2,26E+03	1,15E+06	**Rh114**	6,60E+10	2,90E+10	3,70E+10
				Rh115	3,73E+10	2,77E+09	3,45E+10
Zn72	8,49E+05	8,19E+05	3,04E+04	**Rh116**	2,66E+10	5,30E+09	2,13E+10
Zn73	5,26E+09	2,41E+06	5,25E+09	**Rh117**	2,12E+10	7,81E+06	2,12E+10
Zn74	9,51E+07	2,23E+06	9,29E+07				
Zn76	7,82E+09	2,63E+09	5,19E+09	**Pd112**	7,71E+10	6,93E+10	7,82E+09
Zn77	7,91E+09	1,76E+06	7,90E+09	**Pd113**	8,74E+10	7,68E+10	1,05E+10
Zn78	2,67E+09	1,15E+06	2,67E+09	**Pd114**	1,06E+11	6,60E+10	3,97E+10
Zn79	4,80E+07	4,29E+05	4,76E+07	**Pd115**	6,35E+10	3,73E+10	2,62E+10
Zn80	2,64E+09	4,06E+04	2,64E+09	**Pd116**	7,23E+10	2,66E+10	4,57E+10
Zn81	4,85E+06	4,52E+03	4,84E+06	**Pd117**	4,95E+10	2,12E+10	2,83E+10
				Pd118	4,24E+10	2,70E+09	3,97E+10
Ga74	9,52E+07	9,51E+07	7,29E+04	**Pd119**	8,16E+09	1,61E+07	8,14E+09
Ga75	2,62E+09	2,62E+09	5,61E+05	**Pd120**	1,06E+10	2,06E+06	1,06E+10
Ga76	1,04E+10	7,82E+09	2,63E+09				
Ga77	7,93E+09	7,91E+09	2,02E+07	**Ag116**	9,33E+10	7,23E+10	2,10E+10
Ga78	5,39E+09	2,67E+09	2,72E+09	**Ag117**	5,37E+10	4,95E+10	4,18E+09
Ga79	1,08E+10	4,80E+07	1,08E+10	**Ag118**	6,68E+10	4,24E+10	2,43E+10
Ga80	8,08E+09	2,64E+09	5,44E+09	**Ag119**	6,62E+10	8,16E+09	5,81E+10
Ga81	2,84E+09	4,85E+06	2,84E+09	**Ag120**	4,50E+10	1,06E+10	3,44E+10
Ga82	1,64E+08	1,34E+06	1,62E+08	**Ag121**	5,54E+10	2,66E+09	5,28E+10
Ga83	5,67E+07	1,34E+05	5,66E+07	**Ag122**	1,06E+10	5,99E+06	1,06E+10

Ga84	1,24E+07	9,43E+03	1,24E+07	**Ag123**	7,94E+09	7,71E+05	7,94E+09
				Ag124	2,64E+09	6,13E+04	2,64E+09
Ge78	1,07E+10	5,39E+09	5,29E+09	**Ag125**	5,26E+09	2,98E+03	5,26E+09
Ge79	1,31E+10	1,08E+10	2,22E+09	**Ag126**	1,36E+02	7,02E-20	1,36E+02
Ge80	1,13E+10	8,08E+09	3,23E+09	**Ag127**	1,71E+01	9,64E-07	1,71E+01
Ge81	1,53E+10	2,84E+09	1,25E+10				
Ge82	9,83E+09	1,64E+08	9,66E+09	**Cd118**	9,25E+10	6,68E+10	2,57E+10
Ge83	4,31E+09	5,67E+07	4,26E+09	**Cd119**	7,27E+10	6,62E+10	6,46E+09
Ge84	3,86E+09	1,24E+07	3,84E+09	**Cd120**	7,68E+10	4,50E+10	3,18E+10
Ge85	4,09E+08	1,39E+06	4,08E+08	**Cd121**	1,19E+11	5,54E+10	6,31E+10
				Cd122	6,62E+10	1,06E+10	5,56E+10
As79	1,63E+10	1,31E+10	3,25E+09	**Cd123**	2,15E+10	7,94E+09	1,36E+10
As80	1,40E+10	1,13E+10	2,66E+09	**Cd124**	5,31E+10	2,64E+09	5,05E+10
As81	2,72E+10	1,53E+10	1,19E+10	**Cd125**	1,56E+10	5,26E+09	1,03E+10
As82	2,09E+10	9,83E+09	1,10E+10	**Cd126**	7,89E+09	1,36E+02	7,89E+09
As83	3,95E+10	4,31E+09	3,52E+10	**Cd127**	5,93E+06	1,71E+01	5,93E+06
As84	3,52E+10	3,86E+09	3,14E+10	**Cd128**	1,84E+06	1,02E+00	1,84E+06
As85	2,15E+10	4,09E+08	2,11E+10	**Cd129**	6,87E+05	3,06E-02	6,87E+05
As86	1,12E+10	8,78E+07	1,11E+10	**Cd130**	8,03E+04	1,37E-02	8,03E+04
As87	8,60E+09	5,42E+06	8,60E+09				
				In120	7,94E+10	7,68E+10	2,63E+09
Se84	7,66E+10	3,52E+10	4,14E+10	**In122**	8,20E+10	6,62E+10	1,58E+10
Se85	5,09E+10	2,15E+10	2,94E+10	**In123**	4,56E+10	2,15E+10	2,41E+10
Se86	7,07E+10	1,12E+10	5,95E+10	**In124**	7,95E+10	5,31E+10	2,64E+10
Se87	3,80E+10	8,60E+09	2,94E+10	**In125**	8,20E+10	1,56E+10	6,65E+10
Se88	1,85E+10	2,39E+08	1,83E+10	**In126**	6,07E+10	7,89E+09	5,28E+10
Se89	7,46E+09	4,98E+07	7,41E+09	**In127**	4,52E+10	5,93E+06	4,51E+10
Se90	4,16E+09	1,27E+06	4,16E+09	**In128**	2,68E+10	1,84E+06	2,68E+10
Se91	2,71E+08	2,43E+05	2,70E+08	**In129**	8,85E+09	6,87E+05	8,85E+09
				In130	3,60E+08	8,03E+04	3,60E+08
Br84	7,66E+10	7,66E+10	2,06E+07	**In131**	1,93E+08	4,23E+03	1,93E+08
Br85	5,94E+10	5,09E+10	8,45E+09	**In132**	9,30E+08	2,62E+06	9,28E+08
Br86	9,18E+10	7,07E+10	2,11E+10	**In133**	1,06E+08	1,22E+05	1,06E+08
Br87	8,44E+10	3,80E+10	4,63E+10				
Br88	7,65E+10	1,85E+10	5,80E+10	**Sn127**	6,28E+10	4,52E+10	1,76E+10
Br89	1,07E+11	7,46E+09	9,93E+10	**Sn128**	1,01E+11	2,68E+10	7,47E+10
Br90	4,68E+10	4,16E+09	4,26E+10	**Sn129**	5,18E+10	8,85E+09	4,29E+10
Br91	2,20E+10	2,71E+08	2,17E+10	**Sn130**	3,87E+10	3,60E+08	3,83E+10
Br92	1,09E+10	2,66E+09	8,27E+09	**Sn131**	2,47E+10	1,93E+08	2,46E+10
Br93	3,35E+09	8,23E+05	3,35E+09	**Sn132**	3,33E+10	9,30E+08	3,23E+10
				Sn133	9,75E+09	1,06E+08	9,64E+09
Kr87	9,28E+10	8,44E+10	8,40E+09	**Sn134**	2,62E+09	7,08E+06	2,62E+09
Kr88	1,10E+11	7,65E+10	3,38E+10				
Kr89	1,47E+11	1,07E+11	4,00E+10	**Sb127**	8,52E+10	6,28E+10	2,24E+10
Kr90	1,29E+11	4,68E+10	8,21E+10	**Sb129**	1,09E+11	5,18E+10	5,71E+10
Kr91	1,26E+11	2,20E+10	1,04E+11	**Sb130**	1,03E+11	3,87E+10	6,42E+10
Kr92	1,17E+11	1,09E+10	1,06E+11	**Sb131**	1,31E+11	2,47E+10	1,06E+11
Kr93	4,67E+10	3,35E+09	4,33E+10	**Sb132**	8,37E+10	3,33E+10	5,04E+10
Kr94	3,43E+10	1,14E+08	3,42E+10	**Sb133**	7,84E+10	9,75E+09	6,86E+10
Kr95	4,45E+09	1,64E+06	4,45E+09	**Sb134**	2,58E+10	2,62E+09	2,32E+10
				Sb135	1,49E+10	2,74E+08	1,47E+10
Rb88	1,10E+11	1,10E+11	2,99E+07	**Sb136**	3,45E+09	2,04E+07	3,43E+09
Rb89	1,56E+11	1,47E+11	9,15E+09				

Rb90	1,30E+11	1,29E+11	1,10E+09	**Te132**	2,02E+11	8,37E+10	1,18E+11
Rb91	1,73E+11	1,26E+11	4,65E+10	**Te133**	1,81E+11	7,84E+10	1,03E+11
Rb92	2,10E+11	1,17E+11	9,30E+10	**Te134**	1,85E+11	2,58E+10	1,59E+11
Rb93	1,70E+11	4,67E+10	1,23E+11	**Te135**	9,55E+10	1,49E+10	8,05E+10
Rb94	1,45E+11	3,43E+10	1,11E+11	**Te136**	6,64E+10	3,45E+09	6,29E+10
Rb95	8,25E+10	4,45E+09	7,80E+10	**Te137**	2,30E+10	5,52E+08	2,25E+10
Rb96	4,86E+10	3,12E+09	4,55E+10	**Te138**	7,96E+09	5,84E+07	7,90E+09
Rb97	6,54E+09	1,15E+07	6,53E+09				
Rb98	1,22E+09	3,74E+06	1,21E+09	**I134**	2,54E+11	1,85E+11	6,86E+10
Rb99	2,44E+08	2,78E+05	2,43E+08	**I135**	2,39E+11	9,55E+10	1,44E+11
Rb100	1,79E+07	5,26E+03	1,79E+07	**I136**	1,59E+11	6,64E+10	9,25E+10
Rb101	6,20E+05	3,22E+01	6,20E+05	**I137**	1,53E+11	2,30E+10	1,30E+11
				I138	7,87E+10	7,96E+09	7,07E+10
Sr91	1,75E+11	1,73E+11	2,81E+09	**I139**	4,70E+10	1,74E+09	4,52E+10
Sr92	2,24E+11	2,10E+11	1,43E+10	**I140**	1,54E+10	2,68E+08	1,51E+10
Sr93	2,32E+11	1,70E+11	6,21E+10	**I141**	3,69E+09	7,97E+07	3,61E+09
Sr94	2,38E+11	1,45E+11	9,31E+10				
Sr95	1,70E+11	8,25E+10	8,75E+10	**Xe137**	2,19E+11	1,53E+11	6,69E+10
Sr96	2,34E+11	4,86E+10	1,86E+11	**Xe138**	2,16E+11	7,87E+10	1,37E+11
Sr97	9,07E+10	6,54E+09	8,42E+10	**Xe139**	1,91E+11	4,70E+10	1,44E+11
Sr98	8,10E+10	1,22E+09	7,98E+10	**Xe140**	1,10E+11	1,54E+10	9,42E+10
Sr99	1,95E+10	2,44E+08	1,93E+10	**Xe141**	5,64E+10	3,69E+09	5,27E+10
Sr100	1,23E+10	1,79E+07	1,22E+10	**Xe142**	2,25E+10	6,04E+08	2,19E+10
Sr101	6,90E+08	6,20E+05	6,89E+08	**Xe143**	3,17E+09	1,08E+08	3,06E+09
Sr102	9,02E+07	7,49E+04	9,02E+07	**Xe144**	1,76E+09	1,46E+06	1,76E+09
				Xe145	1,87E+08	1,68E+05	1,87E+08
Y92	2,24E+11	2,24E+11	0,00E+00				
Y93	2,32E+11	2,32E+11	5,61E+07	**Cs138**	2,48E+11	2,16E+11	3,23E+10
Y94	2,47E+11	2,38E+11	8,37E+09	**Cs139**	2,69E+11	1,91E+11	7,86E+10
Y95	1,99E+11	1,70E+11	2,95E+10	**Cs140**	1,91E+11	1,10E+11	8,11E+10
Y96	2,83E+11	2,34E+11	4,83E+10	**Cs141**	1,63E+11	5,64E+10	1,06E+11
Y97	1,85E+11	9,07E+10	9,41E+10	**Cs142**	1,08E+11	2,25E+10	8,53E+10
Y98	1,50E+11	8,10E+10	6,88E+10	**Cs143**	7,74E+10	3,17E+09	7,42E+10
Y99	1,65E+11	1,95E+10	1,46E+11	**Cs144**	3,99E+10	1,76E+09	3,81E+10
Y100	1,04E+11	1,23E+10	9,19E+10	**Cs145**	8,17E+09	1,87E+08	7,98E+09
Y101	6,11E+10	6,90E+08	6,04E+10	**Cs146**	1,70E+09	1,34E+07	1,68E+09
Y102	1,02E+10	9,02E+07	1,01E+10	**Cs147**	2,07E+08	2,37E+05	2,07E+08
				Cs148	3,53E+07	2,62E+04	3,53E+07
Zr97	2,50E+11	1,85E+11	6,56E+10				
Zr98	2,49E+11	1,50E+11	9,93E+10	**Ba141**	2,19E+11	1,63E+11	5,58E+10
Zr99	2,18E+11	1,65E+11	5,28E+10	**Ba142**	1,66E+11	1,08E+11	5,83E+10
Zr100	2,26E+11	1,04E+11	1,21E+11	**Ba143**	1,66E+11	7,74E+10	8,81E+10
Zr101	1,84E+11	6,11E+10	1,23E+11	**Ba144**	1,41E+11	3,99E+10	1,01E+11
Zr102	1,32E+11	1,02E+10	1,22E+11	**Ba145**	6,80E+10	8,17E+09	5,99E+10
Zr103	5,02E+10	1,91E+09	4,83E+10	**Ba146**	4,85E+10	1,70E+09	4,68E+10
Zr104	2,53E+10	2,34E+08	2,51E+10	**Ba147**	1,47E+10	2,07E+08	1,45E+10
Zr105	5,68E+09	5,22E+07	5,63E+09	**Ba148**	8,86E+09	3,53E+07	8,83E+09
				Ba149	6,80E+08	6,11E+05	6,79E+08
Nb98	2,54E+11	2,49E+11	5,29E+09	**Ba150**	7,82E+07	6,92E+04	7,81E+07
Nb100	2,56E+11	2,26E+11	3,03E+10				
Nb101	2,49E+11	1,84E+11	6,51E+10	**La142**	1,66E+11	1,66E+11	7,29E+07
Nb102	1,94E+11	1,32E+11	6,25E+10	**La143**	1,71E+11	1,66E+11	5,90E+09
Nb103	1,79E+11	5,02E+10	1,29E+11	**La144**	1,78E+11	1,41E+11	3,72E+10

Nb104	9,32E+10	2,53E+10	6,79E+10	**La145**	1,33E+11	6,80E+10	6,54E+10
Nb105	8,14E+10	5,68E+09	7,57E+10	**La146**	9,17E+10	4,85E+10	4,33E+10
Nb106	1,64E+10	2,64E+09	1,38E+10	**La147**	9,12E+10	1,47E+10	7,65E+10
Nb107	7,34E+09	2,80E+05	7,34E+09	**La148**	4,56E+10	8,86E+09	3,67E+10
				La149	2,86E+10	6,80E+08	2,79E+10
Mo101	2,54E+11	2,49E+11	5,40E+09	**La150**	3,15E+09	7,82E+07	3,07E+09
Mo102	2,60E+11	1,94E+11	6,63E+10				
Mo103	2,11E+11	1,79E+11	3,16E+10	**Ce145**	1,36E+11	1,33E+11	2,81E+09
Mo104	1,60E+11	9,32E+10	6,68E+10	**Ce146**	1,28E+11	9,17E+10	3,67E+10
Mo105	1,56E+11	8,14E+10	7,44E+10	**Ce147**	1,21E+11	9,12E+10	3,01E+10
Mo106	1,04E+11	1,64E+10	8,79E+10	**Ce148**	8,54E+10	4,56E+10	3,99E+10
Mo107	5,07E+10	7,34E+09	4,34E+10	**Ce149**	6,67E+10	2,86E+10	3,81E+10
Mo108	3,12E+10	2,50E+08	3,09E+10	**Ce150**	3,66E+10	3,15E+09	3,34E+10
Mo109	1,03E+10	2,19E+07	1,03E+10	**Ce151**	1,24E+10	6,44E+08	1,18E+10
Mo110	3,52E+09	1,60E+06	3,52E+09	**Ce152**	1,06E+10	8,38E+07	1,05E+10
Tc103	2,21E+11	2,11E+11	1,05E+10	**Pr148**	1,04E+11	8,54E+10	1,85E+10
Tc104	1,71E+11	1,60E+11	1,07E+10	**Pr149**	7,83E+10	6,67E+10	1,16E+10
Tc105	1,99E+11	1,56E+11	4,32E+10	**Pr150**	6,03E+10	3,66E+10	2,38E+10
Tc106	1,34E+11	1,04E+11	2,93E+10	**Pr151**	4,54E+10	1,24E+10	3,29E+10
Tc107	1,30E+11	5,07E+10	7,98E+10	**Pr152**	2,52E+10	1,06E+10	1,46E+10
Tc108	7,17E+10	3,12E+10	4,05E+10	**Pr153**	2,01E+10	8,10E+08	1,93E+10
Tc109	6,14E+10	1,03E+10	5,11E+10	**Pr154**	4,01E+09	1,15E+08	3,89E+09
Tc110	2,56E+10	3,52E+09	2,21E+10				
Tc111	1,40E+10	2,50E+08	1,37E+10	**Nd152**	3,13E+10	2,52E+10	6,03E+09
Tc112	3,68E+08	3,22E+07	3,36E+08	**Nd153**	2,96E+10	2,01E+10	9,49E+09
Tc113	2,74E+09	2,42E+06	2,74E+09	**Nd154**	1,09E+10	4,01E+09	6,91E+09
				Nd155	9,60E+09	3,07E+09	6,53E+09
				Nd156	6,00E+09	7,62E+07	5,93E+09

Annex 10

10 Diffusion-effusion efficiency and expected radioactive beam intensities:

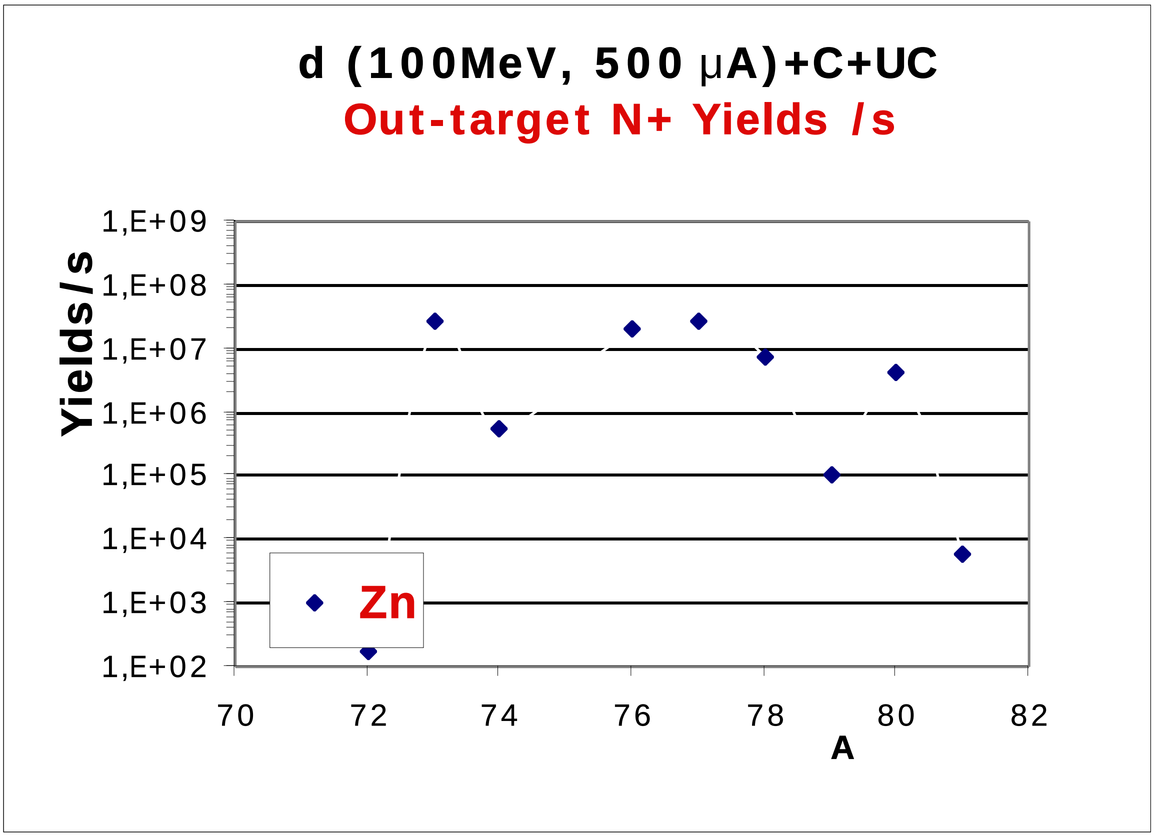
Critical steps for the radioactive beam production by the ISOL method are mainly the diffusion in the target and effusion between target and ion-source.

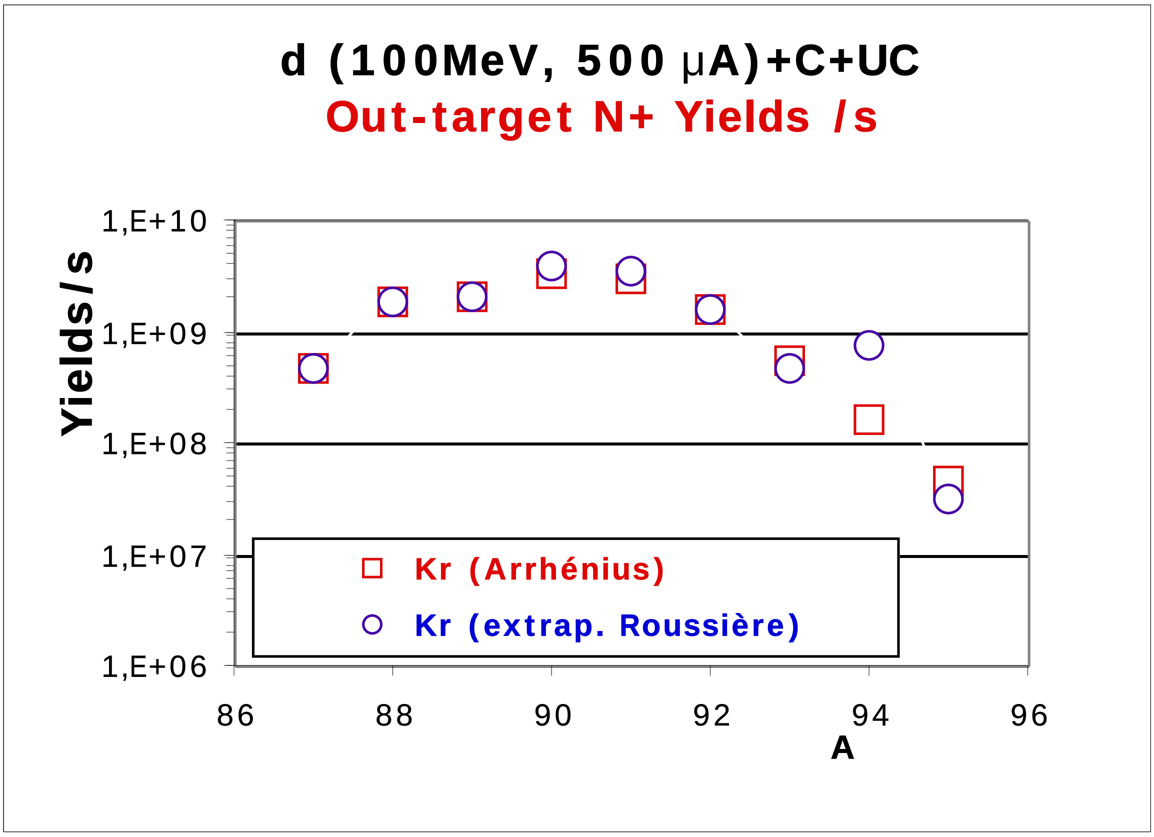
Despite a large experimental and theoretical effort, the behavior of such ions implanted into materials and the details of their thermal transport or their trapping remain incompletely understood. The Arrhenius diffusion coefficients are measured for numerous elements mainly in W, Ta, Re or C-matrix and for noble gaseous elements in a C-matrix. To our knowledge, these coefficients are not known for different tracers in uranium carbide matrices of different densities. A European RTD project “TARGISOL” N° HPRI-2001-50063 has been proposed and accepted in order to achieve progress in this critical field. However, we cannot construct a new machine without determining the expected radioactive beam intensities. Two approaches were realized:

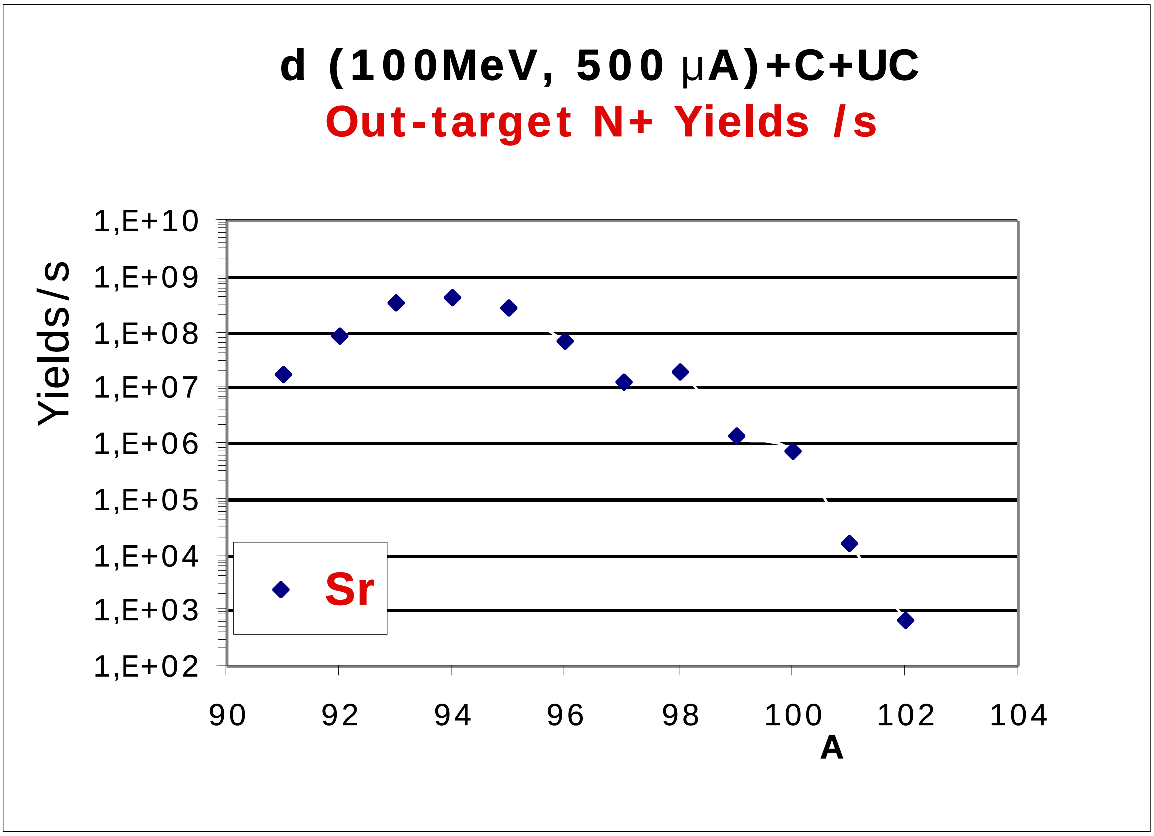
The first one is based on a comparison of the predicted in-target production yields (using the Ficner code and the known cross-sections) and those measured after diffusion-effusion and ionization in the PARRNe2 experiment. This work, done by the Orsay groups, gives a good indication of the diffusion efficiency for different elements in an uranium carbide target and using a ion transfer pipe of small diameter as realised at PARRNe.

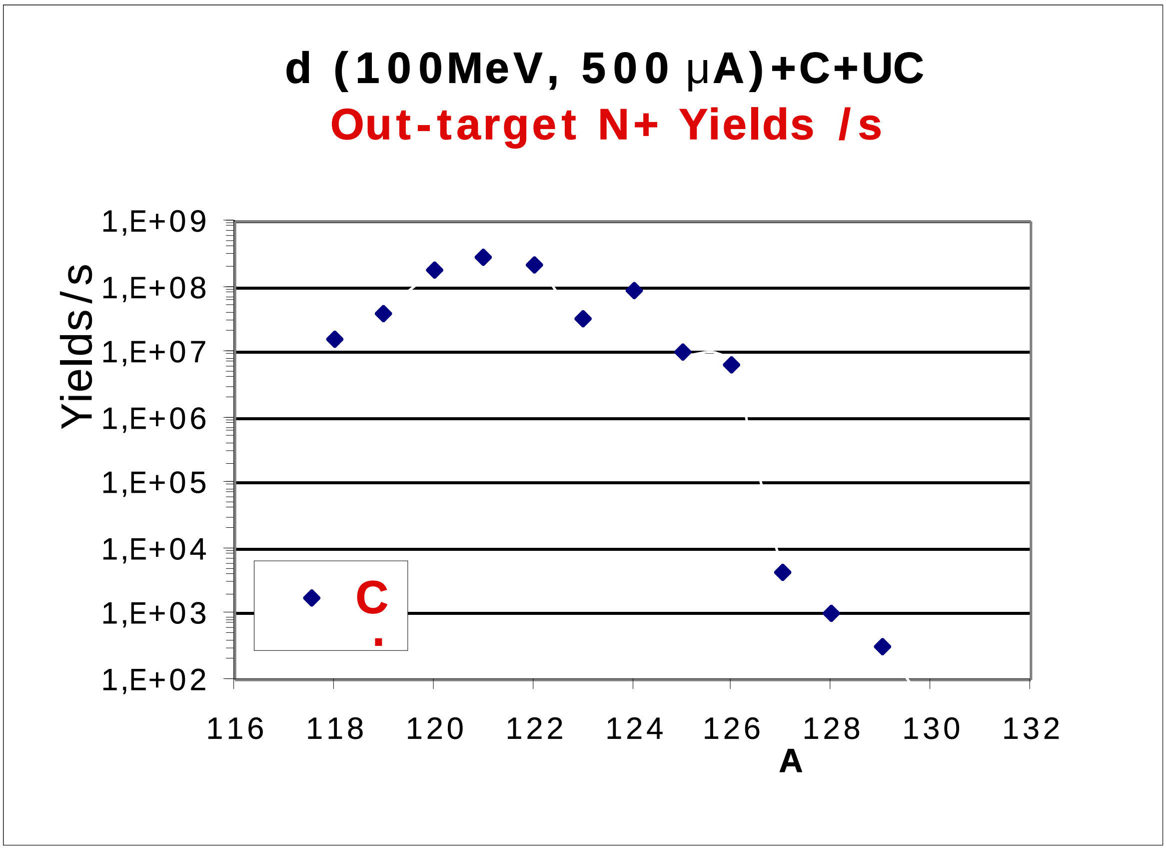
The second one, performed at GANIL and based on a theoretical simulation of the diffusion-effusion process, provides some idea of the influence of the target-source geometry. However, since the diffusion-effusion parameters for uranium carbide are not known as mentioned above, diffusion-effusion parameters for a C or Ta-matrice are used instead.

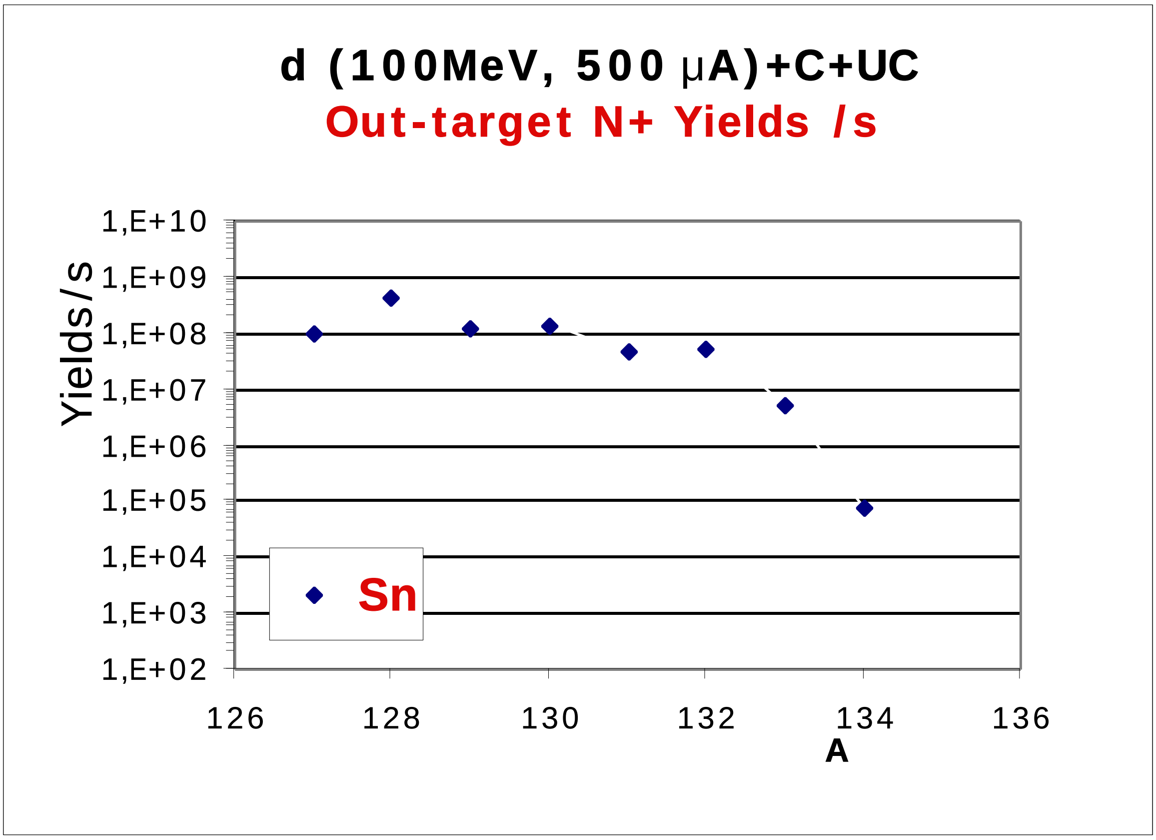
The expected radioactive beam intensities (after diffusion, effusion, ionization and acceleration) are shown in the following plots for some elements: Kr, Xe, I, and Cd, using the first method quoted and for Zn, Kr, Sr, Sn Sb and Xe using the second method. The experimental configuration is the same as in annex 9. We use a deuteron (100 MeV, 500 microA) beam onto a thick C-converter. The target is composed of uranium carbide (density 2.3 g/cm3, C:U=9:1) placed at 2 cm behind the converter. The in-target production yields are calculated using the LAHET-MCNP-CINDER codes. The beam width is 4 cm onto a 6 cm target diameter. The target is composed of 56 slices of 1 mm thickness spaced by 0.5 mm. The assumed 1^+ and $1^+/N^+$ ionization efficiencies are the same 90% (1^+) and 12% ($1^+/N^+$) for Kr and Xe, 30% (1^+) and 4% ($1^+/N^+$) for Zn, Sr, Sn, I and Cd. The assumed transfer efficiency is 50% in the CIME cyclotron.

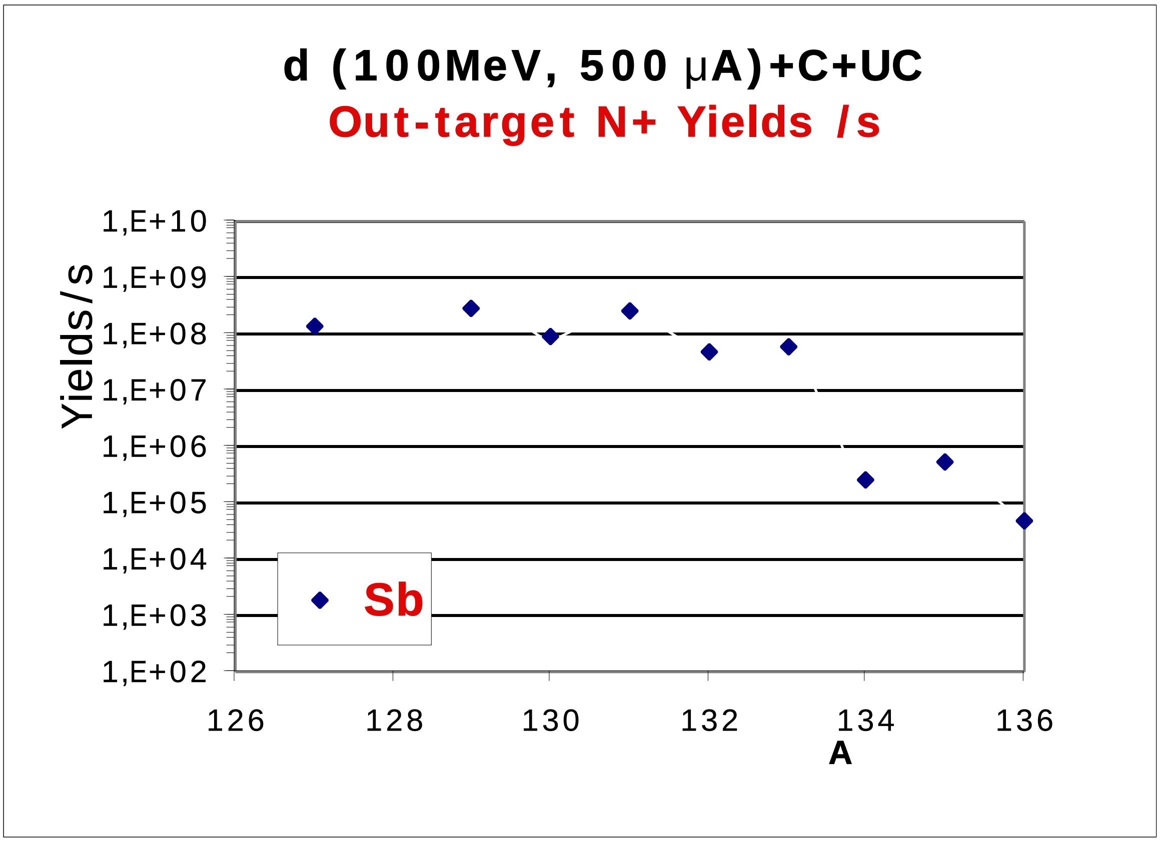

d (100MeV, 500 µA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
Zn
70
72
74
76
78
80
82
A

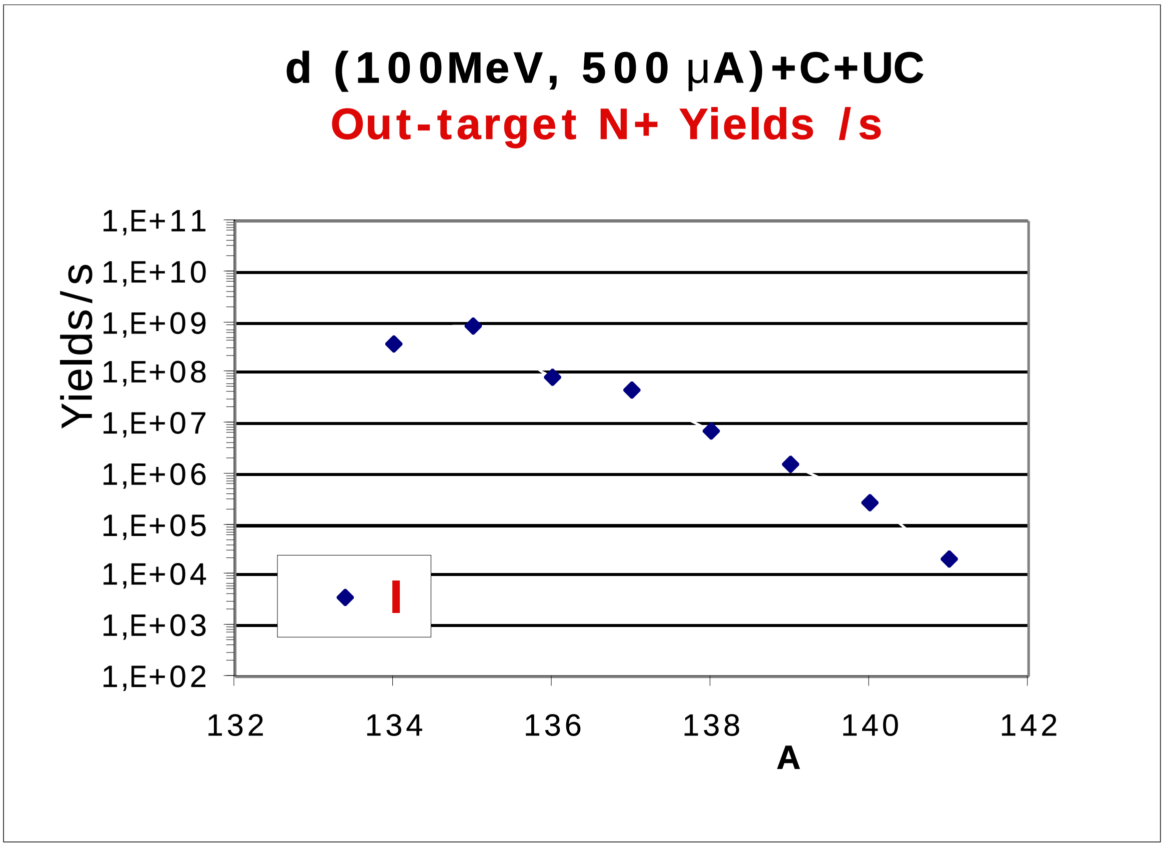

d (100MeV, 500 µA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
Kr (Arrhénius)
Kr (extrap. Roussière)
86
88
90
92
94
96
A

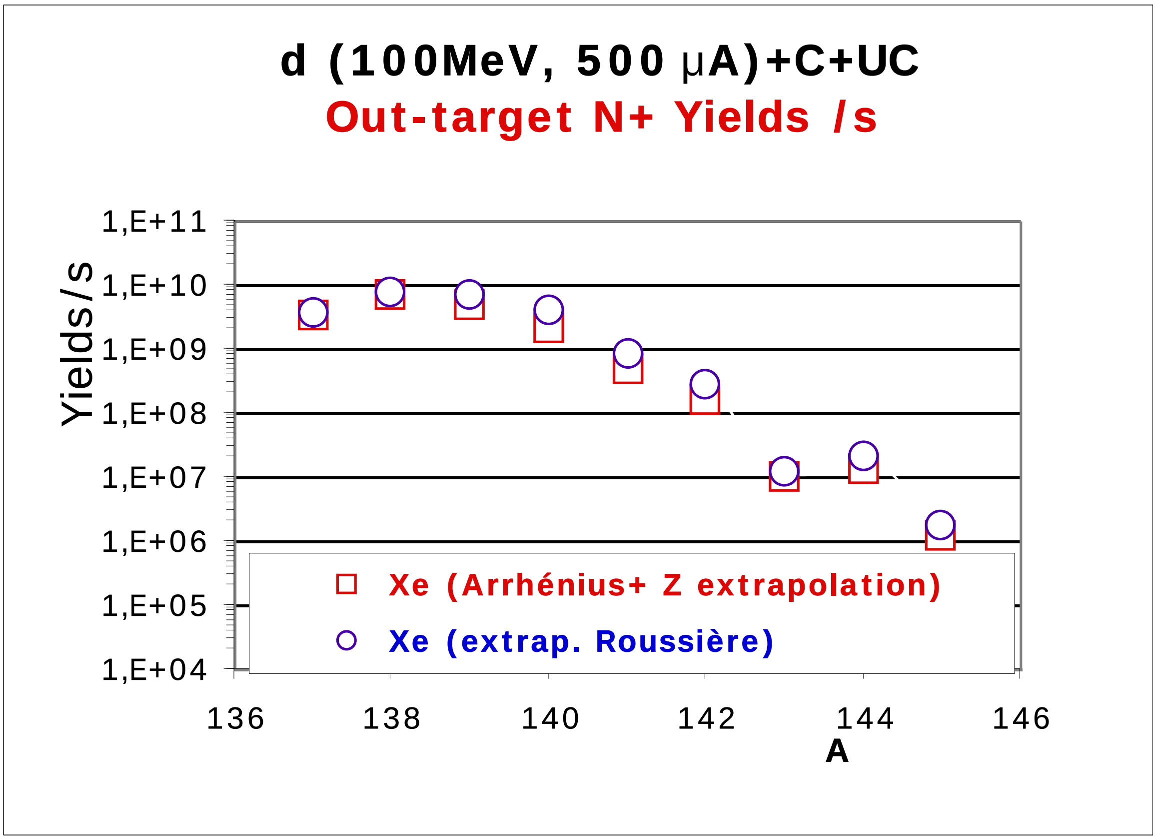

d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
90
92
94
96
98
100
102
104
A
Sr


d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
116
118
120
122
124
126
128
130
132
A
C


d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
Sn
126
128
130
132
134
136
A


d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
Sb
126
128
130
132
134
136
A


d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+11
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
1,E+03
1,E+02
132
134
136
138
140
142
A


d (100MeV, 500 μA)+C+UC
Out-target N+ Yields /s
Yields/s
1,E+11
1,E+10
1,E+09
1,E+08
1,E+07
1,E+06
1,E+05
1,E+04
Xe (Arrhénius+ Z extrapolation)
Xe (extrap. Roussière)
136
138
140
142
144
146
A

Annex 11

11. Implantation Proposal

<u>45 MeV Electron Option:</u>

The scheme n°1 shows the implantation proposed for the electron option. The different parts of the facility are the following :

1. Linear accelerator in a tunnel. The electron energy, of 45 MeV in a first step, can be increased by increasing the cavity field. A 2 m thick roof covers the tunnel.
2. Linac gallery where the electronic devices (RF devices in particular) are installed.
3. Transit gallery to access to the linac gallery and to the LIRAT experimental area from outside. A crane is used to handle the equipment.
4. Electron beam dump.
5. Target and mono-charge state source at a bottom of a kind of well. The beam line is oriented vertically.
6. Secondary well to handle and extract the target after irradiation.
7. Mass separator consisting in a low magnetic field dipole.
8. Free room for very high efficiency mass spectrometer.
9. Charge breeder.
10. Separator secondary output to provide the low energy experiments (LIRAT) with species not used by CIME.
11. Junction with the CIME injection line . The identification bench is adapted to be used jointly by SPIRAL I and II.
12. Existing SPIRAL cave 1
13. SPIRAL cave 2 not yet equipped. Probably this cave will be equipped with a two-stage ionisation system.
14. Low energy experiment area (LIRAT)
15. Free room to install a booster to post-accelerate the beam coming from CIME or directly from the target via a charge breeder.
16. Beam line to possibly transport ions from SPIRAL target to GANIL injector or from CIME to CSS2 in order to re-accelerate them.
17. Limit of an external building for services. This building could be an extension of the present SPIRAL building.

<u>80 MeV Deuteron Option:</u>

The scheme n°2 shows the implantation adapted to the 80 MeV deuteron option:

1. Cyclotron cave. The deuteron source is installed above the cyclotron.
2. Gallery where the electronic devices (RF devices and power converters in particular) are installed.
3. Transit gallery to access to the linac gallery and to the LIRAT experiment area from outside. A crane is used to handle the equipment.
4. -

5.-17 see electron option

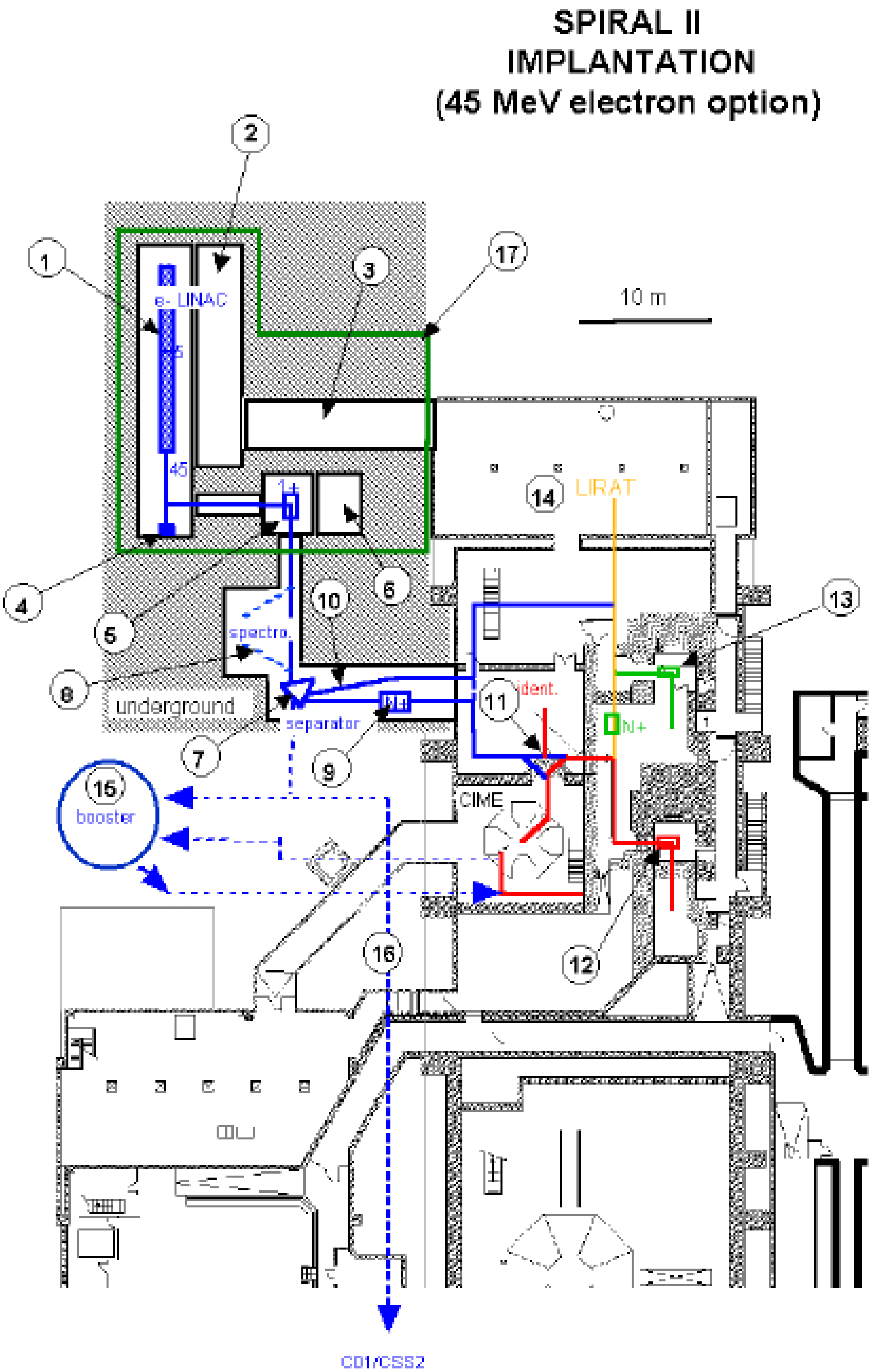
<u>40 MeV Deuteron Option:</u>

The scheme n°3 shows the implantation adapted to the 45 MeV deuteron option :

1. RFQ tunnel.
2. Linac tunnel. The output energy of the linac can be increased by extending the tunnel to the nort
3. Gallery where the electronic devices (RF devices and power converters in particular) and the cryogenic plant are installed
4. Deuteron beam dump

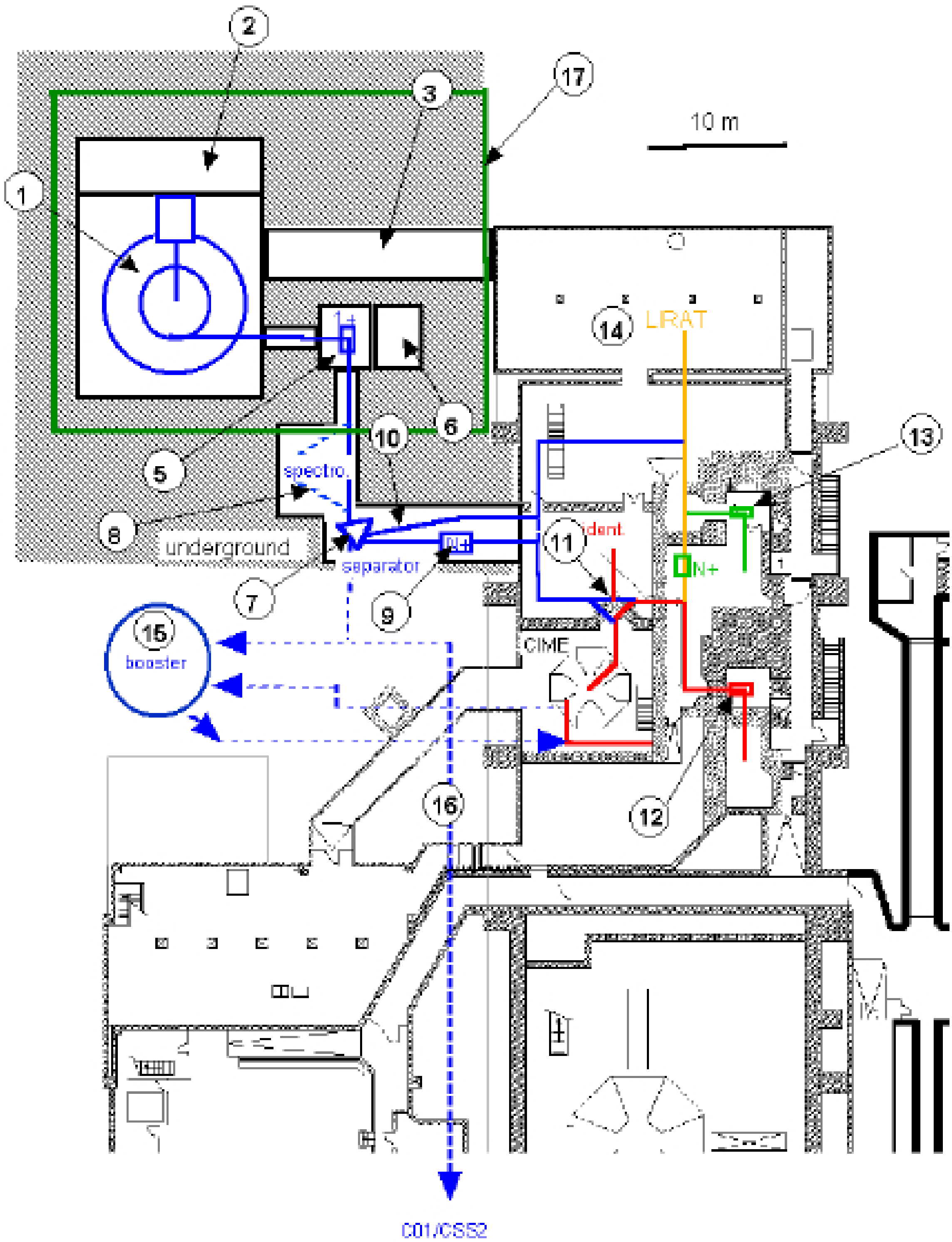
5..-17 see electron option

18. Second production cave (future extension)

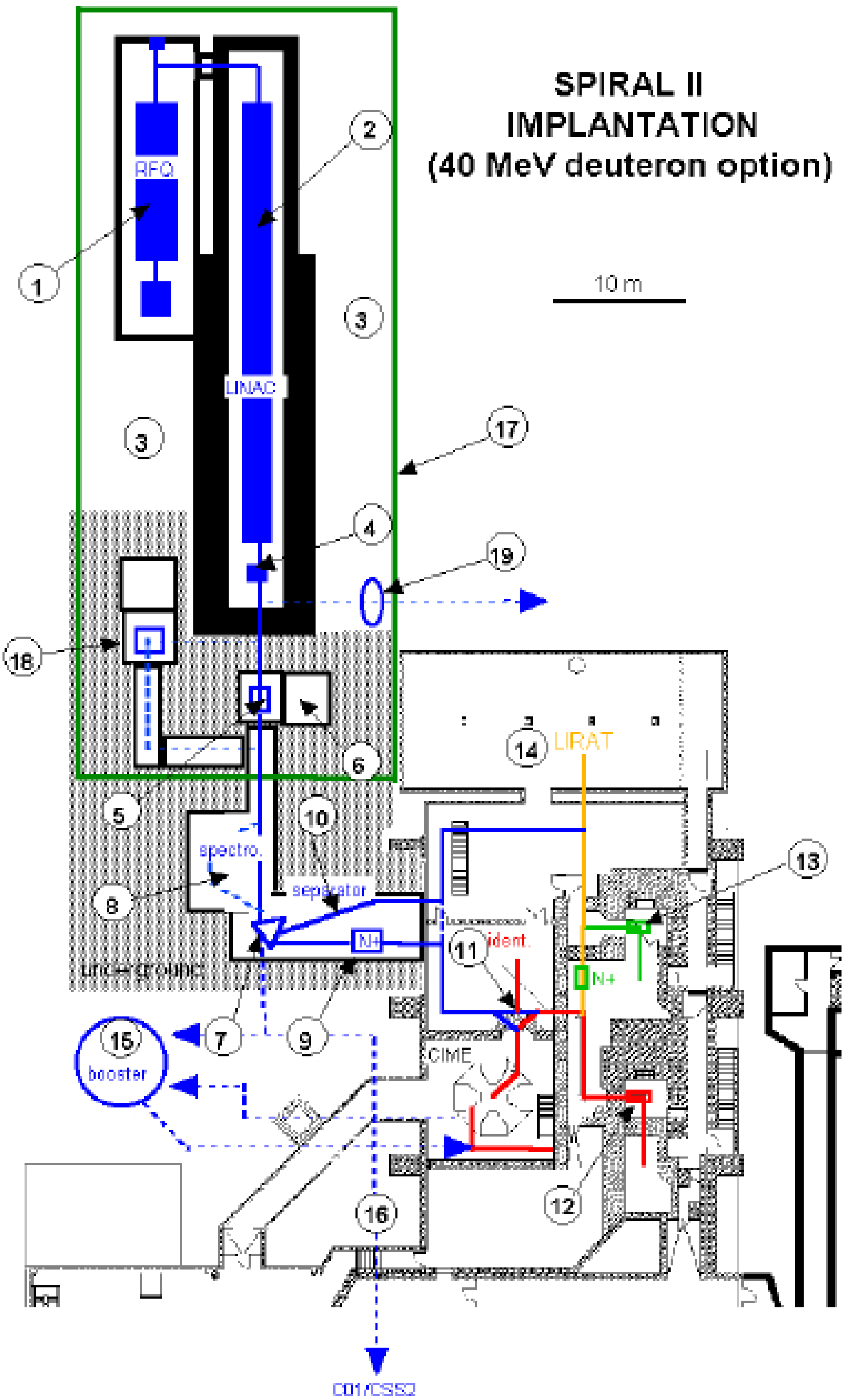

SPIRAL II
IMPLANTATION
(45 MeV electron option)
10 m
e- LINAC
45
1+
LIRAT
spectro.
underground
separator
N+
ident.
CIME
booster
CD1/CSS2
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17

Scheme 1

SPIRAL II IMPLANTATION (80 MeV deuteron option)



Scheme 2


SPIRAL II
IMPLANTATION
(40 MeV deuteron option)
10 m
RFQ
LINAC
spectro.
separator
N+
ident.
LIRAT
CIME
booster
underground
C01/CSS2
1
2
3
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19

Scheme 3