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A new method for measuring the absolute efficiency of Isotope Separation On-Line (ISOL) systems is described. The overall efficiency is deduced from the ratio between the rate of the radioactive beam detected after analysis and the rate of incident radioactive beam in the target. It is shown that the unfolding of the different efficiencies, i.e., diffusion, effusion, ionisation and transport allows us to deduce the diffusion constants when the absolute efficiency data is obtained as a function of the target temperature. Using this technique we extracted the fundamental Arrhenius constants for diffusion of Ar and Ne in Carbon targets used in the SIRa experiments at GANIL.

1. INTRODUCTION

The study of release properties of a target and the total efficiency of Isotope Separation On-Line systems (ISOL) [1] is of paramount importance in producing a radioactive ion beam (RIB). For the SPIRAL [2] project at GANIL, France, the properties of the target used in the production of radioactive elements and the performance of the ECRIS (Electron-Cyclotron Resonance Ion Source) [3] on line are being studied using the SIRa (Radioactive Ions Separator) [4] separator. For this purpose we developed a new technique in order to deduce the overall absolute efficiency of the separator and, in particular, the release properties of the target.

This technique is used for measuring the overall efficiency of the SIRa separator with radioactive ^{35}Ar ($T_{1/2} = 1.77\text{s}$), ^{19}Ne ($T_{1/2} = 17.22\text{s}$) and ^{18}Ne ($T_{1/2} = 1.67\text{s}$) atoms in a Carbon target. Measurements of this efficiency as a function of the target temperature allowed us to deduce the fundamental constants of diffusion for Ar and Ne gas.

Section 2 describes the method, the separator SIRa, the geometry of the target-ECRIS system and the experiment details of this new method. Section 3 describes the measurements of the different contributions of the total efficiency of the separator. Section 4 presents the theoretical aspects of the diffusion and effusion, a description of the proposed method for extracting the Arrhenius constants and the analysis of the results. Section 5 gives the summary and conclusions.

2. METHOD FOR MEASURING THE OVERALL EFFICIENCY OF THE SEPARATOR.

2.1. Description of the method.

The aim of the method remains in the use of the facility of the GANIL accelerators to produce by projectile fragmentation a secondary radioactive beam and to implant it in the target of the ISOL system. Therefore, the overall efficiency can be extracted from the ratio between the detected rate at the end of the separator and the implanted rate in the target. This efficiency is intrinsically dependent of the transit time of the radioactive probe. It is important to note that around 100A MeV the total probability of reaction of a beam in an infinite target is of the order of 1%, which is negligible in view of the precision required.

2.2. The SIRa Separator and the target-ECRIS system.

The SIRa separator is usually used to produce radioactive multicharged ion beams by projectile fragmentation of a stable primary heavy ion beam in a thick target. The beam

can have an energy up to 100 A.MeV with a power up to 400 W. The radioactive produced elements diffuse out of the target, go through the transfer tube, and arrive into the chamber of an ECRIS where they are ionised. After extraction, the radioactive ions are selected and transported through the separator to a detection point.

Two different set-ups have been used for the efficiency measurements. The first one consisted of an external cylindrical carbon target (made with 30 slices 0.7mm thick and 0.3mm spaced) linked to the ECR3 ion source [5] by a 60cm long tantalum transfer tube. The carbon of the target was furnished by Le Carbone Lorraine, France (grade 2318) with a grain size of 4 μ m and 8% open porosity. With this set up, we measured the overall efficiency of SIRa with the radioactive ^{35}Ar and ^{18}Ne elements.

In the second set-up, we used the new internal target ion source system called SHyPIE [6] for measuring the overall efficiency with the ^{19}Ne . A pyrolytical carbon target block was placed inside the ion source, just behind the plasma zone.

2.3. Experimental details.

In order to measure the overall efficiency of SIRa in an equivalent production condition, a high energy secondary beam is required. The secondary radioactive beam of ^{35}Ar , ^{19}Ne or ^{18}Ne has been produced by projectile fragmentation of respectively ^{36}Ar , ^{24}Mg and ^{20}Ne primary beams at $E = 95\text{A MeV}$ in a target placed between the solenoids of SISSI [7] and was selected by the α -shaped spectrometer of GANIL operated as a recoil separator as shown schematically in the figure 1. The secondary RIB rate was determined using a surface barrier Si detector 300 mm thick, located in front of the SIRa target. The identification of the secondary RIB is unambiguously made by energy loss in the Si detector and the time of flight between the production target and the detector. The ^{35}Ar , ^{19}Ne or ^{18}Ne radioactive atoms are implanted and released by the target at a high temperature (between 1500 K and 2000 K), effuses threw to the ECR source, are ionised, extracted, selected by the separator and implanted in a plastic tape located outside the experimental vault of SIRa itself. The γ -rays emitted by the decay of the radioactive nuclei were detected using a Ge detector located just behind the tape. The overall efficiency of the separator is defined as the following equation:

$$\varepsilon_t = \frac{N_s}{N_i} \quad (1)$$

where N_s and N_i are respectively the number of ions per second implanted in the plastic tape and the number of ions per second implanted in the target. The determination of N_s for all cases - ^{35}Ar , ^{19}Ne or ^{18}Ne - was also unambiguous due to the existence of known γ -rays with tabulated branching ratios.

3. MEASUREMENT OF THE DIFFERENT EFFICIENCIES OF THE SEPARATOR.

The overall efficiency of the system can be expressed by the following relationship:

$$\varepsilon_t = \varepsilon_\mu \varepsilon_v \varepsilon_{ts} \varepsilon_i \varepsilon_{tr} \quad (2)$$

where ε_μ is the diffusion efficiency out of the target, ε_v is the effusion efficiency in the transfer tube, ε_{ts} is the geometrical transfer efficiency, which is essentially associated with leaks on the transfer between the target container and the ion source, ε_i is the ionisation efficiency and ε_{tr} is the beam transport efficiency. The product $\varepsilon_{ts} \varepsilon_i \varepsilon_{tr}$ could be determined off-line, independently of the radioactive probe nuclei due to the fact that they are only function of the atomic/chemical properties of the selected element. For this purpose a calibrated leak of ^{40}Ar or ^{22}Ne gas has been injected continuously in the target container during the irradiation. Here we note that the unfolding of each particular efficiency - ε_{ts} , ε_i , ε_{tr} - is not a requirement for deducing the $\varepsilon_\mu \varepsilon_v$ product from the experimental ε_t data, as we will see in the following section. Nevertheless, each efficiency can be independently deduced as the following:

3.1. Transport.

The transport efficiency ε_{tr} has been calculated by the ratio between the integral of the full beam arriving at the faraday cup placed in front of the plastic tape, downstream the separator, and the current flux of the High Voltage power supply of beam extraction, deducing losses already measured. This efficiency has also been verified using a faraday cup placed just after the extraction electrode. This faraday cup can measure the full extracted beam current in cases where the emittance of the ion source is small. It has been done off-line, tuning the ECRIS for low total extraction current. The agreement between the measurement using the faraday cup and the current flux of the power supply was better then 5%.

3.2. Ionisation.

The ion source efficiency ε_i can be described as the product between an ionisation and a delay efficiency, where the latter represents the time spent by an atom inside the source before being extracted. For noble gases with $T_{1/2} > 100\text{ms}$, the sticking time on the walls is negligible and the confinement time is much smaller then the life time of the radioactive element. In this case the ion source efficiency can be assumed to be the same for stable and radioactive atoms. Experimentally, the ionisation efficiency has been calculated by the ratio between the current of the different charge states of ^{40}Ar or ^{22}Ne

detected in the faraday cup placed in front of the plastic tape and the calibrated leak placed directly in contact with the ECRIS chamber - exactly in the H.F. coaxial connector - corrected for the transport efficiency.

3.3. Transfer between target container and ion source

The transfer efficiency ϵ_{ts} - between target container and ion source - has been determined, in a first step, by measuring the flux of the calibrated leak of ^{40}Ar or ^{22}Ne gas threw to the ion source using a calibrated leak detector. It has also been verified measuring the leak of the same gas to outside of the system. This technique gives an experimental error of the order of 20% in the value of ϵ_{tr} . Another method, which gives better precision, was to place the same calibrated leak in contact with the target container and measure the full ^{40}Ar or ^{22}Ne beam detected in the faraday cup placed in front of the plastic tape of SIRa. The ratio between the measured intensity of beam and the beam observed with the leak placed at the H.F. coaxial connector gives the transfer efficiency. This method gives a precision of the order of 5%. The value of ϵ_{tr} can be already verified in the on-line run, as will be discussed below.

Finally, the diffusion-effusion efficiency can be deduced after measuring the overall efficiency of the separator and the transfer, ionisation and transport efficiencies. This will allow to study the release properties of different targets with different incident beams.

4. ANALYSIS AND RESULTS.

4.1. Mesured efficiencies.

The results obtained for the transport efficiencies are quite different for the experiment with ^{18}Ne and ^{35}Ar . During the experiment using ^{35}Ar as probe radioactive element we had an important leak between the connection of the target container and the transfer tube. Therefore, we obtained for ^{35}Ar $\epsilon_{ts} = 0.093$ (0.025). For the ^{18}Ne experiment, the problem has been partially solved and the efficiency was $\epsilon_{ts} = 0.50$ (0.03). In the experiment using ^{19}Ne , the target was inside the source, therefore $\epsilon_{ts} = 1.0$ by definition.

The ionisation efficiencies obtained for all cases were excellent. The ionisation efficiencies were $\epsilon_i = 0.16$ (0.01) for ^{35}Ar in the charge state 8+, $\epsilon_i = 0.19$ (0.01) for ^{18}Ne in the charge state 5+ and $\epsilon_i = 0.15$ (0.01) for ^{19}Ne in the charge state 4+.

The transport efficiency between the ion source and the plastic tape was dominated by the large divergence of the extracted beam, which was estimated to be larger than 50 mrad. Therefore, we had important losses of beam due to the transport. The transport efficiencies were $\epsilon_{tr} = 0.23$ (0.01) for the ^{35}Ar beam, $\epsilon_{tr} = 0.36$ (0.03) for ^{18}Ne and $\epsilon_{tr} = 0.25$ (0.01) for ^{19}Ne .

The product of these efficiencies independent of life-time were:

- i) for ^{35}Ar : $\varepsilon_{ts}\varepsilon_i\varepsilon_{tr} = 0.0034$ (0.0010)
- ii) for ^{18}Ne : $\varepsilon_{ts}\varepsilon_i\varepsilon_{tr} = 0.034$ (0.003)
- iii) for ^{19}Ne : $\varepsilon_i\varepsilon_{tr} = 0.037$ (0.001)

4.2. Theory of diffusion and effusion

The diffusion of an element in a solid state material is governed by the Fick's laws, which has been solved already for several conditions [8-10]. In the case of diffusion of a radioactive element in a porous material the diffusion equation should include a term which considers the fact that the probe element can decay during the diffusion. However, after releasing from the target material, the radioactive element is conducted up to the ion source by a transfer tube. The mean time spent on the transit from the target to the ion source is the sum between the mean time of flight from the target to the ion source and the mean total sticking time in the walls of the container and transfer tube. Considering χ as being the mean number of collisions with the surface of the container and the transfer tube, τ_a as the mean sticking time per collision and τ_f the mean time of flight between two successive collisions, we can deduce that the mean total effusion time is equal to:

$$\tau_v = \chi(\tau_a + \tau_f) \quad (3)$$

The effusion efficiency can be written as:

$$\varepsilon_v(\lambda) = v / (v + \lambda) \quad (4)$$

$$\text{where } v = 1 / \tau_v \quad (5)$$

Finally, for the ^{35}Ar and ^{18}Ne experiments, the product of the diffusion efficiency out of the $4\mu\text{m}$ grain size carbon target for a spherical symmetry and the effusion efficiency in the transfer tube can be written as [8,10]:

$$\varepsilon_\mu \varepsilon_v(\lambda) = \frac{3(\sqrt{\pi^2 \lambda / \mu_0} \coth \sqrt{\pi^2 \lambda / \mu_0} - 1)}{(1 + \lambda/v) \pi^2 \lambda / \mu_0} \quad (6)$$

where $\lambda = \ln 2 / T_{1/2}$, $T_{1/2}$ is the half life of the radioactive probe nucleus,

$$\mu_0 = \pi^2 D / d^2 \quad (7)$$

$$\text{and } D = D_0 e^{(-E_A / kT)} \quad (8)$$

where d and T are the grains radius and the temperature of the carbon target, D_0 and E_A are the Arrhenius coefficients.

For the ^{19}Ne experiment, there is no effusion efficiency and the diffusion efficiency out of the pyrolytical target, assumed as an infinite foil of thickness d , can be written as [8,10]:

$$\varepsilon_{\mu}(\lambda) = \frac{\tanh \sqrt{\pi^2 \lambda / 4 \mu_0}}{\sqrt{\pi^2 \lambda / 4 \mu_0}} \quad (9)$$

The Arrhenius coefficients D_0 and E_A can be easily deduced if we measure $\varepsilon_{\mu}(\lambda)$ or $\varepsilon_{\mu}\varepsilon_v(\lambda)$ as a function of the target temperature.

4.3. Results and extrapolations

The diffusion efficiencies, deduced from the overall efficiencies, as a function of the target temperature are shown in figure 2, for the noble gases radioactive ions of ^{35}Ar and ^{18}Ne releasing from a $4\mu\text{m}$ grain size carbon target and for ^{19}Ne releasing from a pyrolytical target.

The fit of the experimental results, using the equation (8) for the powder target and the equation (9) for the pyrolytical one, have given the following Arrhenius coefficients :

- i) for ^{35}Ar : $\log(D_0/d^2) = 3.8 (0.5) \text{ s}^{-1}$ and $E_A = 55.0 (4.4) \text{ kcal/mol}$, $\chi^2 = 0.55$
- ii) for ^{18}Ne : $\log(D_0/d^2) = 1.04 (0.40) \text{ s}^{-1}$ and $E_A = 28.7 (2.9) \text{ kcal/mol}$, $\chi^2 = 1.52$
- iii) for ^{19}Ne : $\log(D_0/d^2) = 1.55 (0.50) \text{ s}^{-1}$ and $E_A = 37.7 (4.4) \text{ kcal/mol}$, $\chi^2 = 0.57$

These values are given for the particular carbon and target geometries used in this experiment. They can not be used as universal carbon diffusion coefficients [11].

However, knowing the Arrhenius coefficients for one given nucleus-target couple, it is possible to extrapolate the diffusion efficiency for the other isotopes of this measured nucleus. The extrapolations of the diffusion efficiencies as a function of the half life, which represents the different isotopes, are shown in figure 3.

First one can conclude that the diffusion out of a $4\mu\text{m}$ grain size carbon target is very similar for Ne and Ar elements. Second, the powder graphitized target has a better diffusion property when compared with the pyrolytical one. This kind of information is very important for choosing the target which will allow the better releasing of the produced radioactive elements. This is particularly important for the SPIRAL project at GANIL.

5. SUMMARY.

We present a new method for measuring the absolute overall efficiency of Isotope Separation On-Line systems.

Based on the on-line measurements of the absolute efficiency of the SIRa separator as a function of the target temperature, we deduced the Arrhenius coefficients for ^{35}Ar , ^{18}Ne and ^{19}Ne . This technique can be, in principle, applied for any target-ion source system for on-line radioactive beam production.

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Fig. 1. Schematic view of the GANIL accelerators and the SIRa experimental cave.

Fig. 2. Diffusion efficiency as a function of the target temperature for ^{35}Ar and ^{18}Ne releasing from a powder carbon target and for ^{19}Ne releasing from a pyrolitical carbon target. The points correspond to the experimental data and the lines correspond to fits using the Arrhenius coefficients found in the text.

Fig. 3. Diffusion efficiency as a function of the half life for Ar and Ne isotopes releasing from a powder carbon target and for Ne isotopes releasing from a pyrolitical carbon target.

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