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Adsorption of radioactive ions on carnauba-wax aerosols

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Abstract. A new method based on parallel aerosol size spectrometry and γ-spectrometry is introduced for the measurement of short-lived radioactive ions, fission products or super-heavy elements produced at accelerators. Furthermore a new aerosol generator is presented.

The possibility of controlling and changing the aerosol size distribution in the helium aerosol jet produced by the aerosol generator allows the process of the adsorption and transport of radioactive ions on aerosols to be examined for the first time. This is due to the fact that the distribution is surveyed on-line using a negligible part of its total volume and parallel to the transporting flow. The radioactivity of the transported ions is measured by a germanium detector in offline position. In principle, both an on- or offline position with narrow multi-detector geometry (e.g. $\beta\gamma\gamma$) is possible.

PACS. 42.68.Jg Effects of aerosols – 07.77.Ka Charged-particle beam sources and detectors

1 Introduction

The study of the decay properties of fission isomers and short-lived isotopes approaching the r-process path requires a very fast transportation system because of decreasing half-lives. Therefore a new and highly effective collection system has to be invented, since the production rate of the very neutron-rich isotopes decreases with increasing neutron excess, too [1].

A new aerosol generator based upon the use of carnauba wax, had originally been developed for the collection of all decay products in the radon reference chamber of the PTB. Carnauba-wax aerosols have unique physical and chemical properties as regards their form, size, density, acidic value etc. It mainly consists of carbon-hydrogen molecules with a preferred chain length of 58. Molecular dynamics simulation and pictures taken by an electronic microscope proved, that these carnauba wax chain molecules produce nearly ideal spheres under well-defined conditions [2].

The aerosol generator was redesigned to optimize the collection efficiency for the fast transport of short-lived radioactive ions (e.g. fission products). The mean diameter of aerosols may be varied from 10 nm to 800 nm; moreover, the aerosol size distribution and concentration can be kept constant over several days, very easily. In a first test run, the carnauba-wax aerosol generator was successfully installed together with the aerosol size spectrometer and a γ-detector in offline position at the 'Institut des Sciences Nucléaires' (ISN, Grenoble, France), using a ^{252}Cf-sf source. The results obtained by this new method are now applied and verified at the LOHENGRIN mass separator of the 'Institut Max von Laue Paul Langevin' (ILL, Grenoble, France) for the measurement of short-lived neutron-rich isotopes. A overall collection efficiency of (95 ± 3.5) % was already obtained at LOHENGRIN.

2 Experimental method

The production of aerosols is based on the method of the condensation of vapour at a well-defined temperature [3], [4], by which aerosols of different sizes and concentrations can be obtained [5], [6].

Carnauba wax [7] is put into a sample boat of elliptical shape connected to a condensation volume and heated by an insulated wire cord by which it is surrounded (see Fig. 1). The critical nuclei and the aerosols grown from these critical nuclei are formed from the vapour.

To obtain data for the adsorption process of radioactive ions on aerosols a highly efficient source of different elements, isotopes and ionic charges is needed. Therefore, a spontaneous fission source (^{252}Cf, A = 7.0 MBq) is chosen. The aerosol generator, presented here, was applied successfully (together with an aerosol measurement system, a germanium detector for high-resolution γ-spectrometry and the ^{252}Cf-sf source) for the measurement of the adsorption and transport of fission products on carnauba-wax aerosols [8] for the first time.

The fission products of the ^{252}Cf-sf source are transported by the helium aerosol jet to a germanium detector in offline position using a collection tape. Parallel to γ-spectrometry, the aerosol production is surveyed by aerosol size spectrometry using a negligible part of the volume.

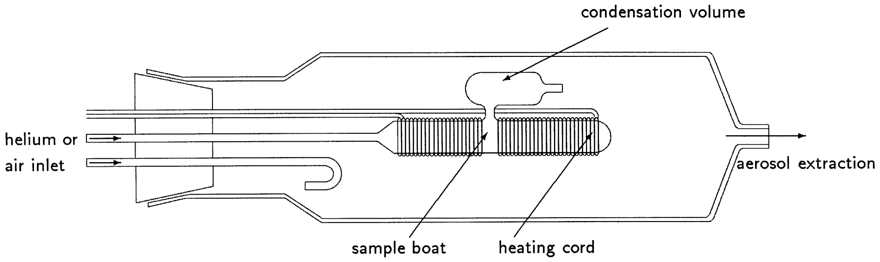

Fig. 1. Longitudinal section of the aerosol generator. The mounted condensation volume provides a well-directed and highly concentrated aerosol jet of great stability in time and in aerosol size distribution

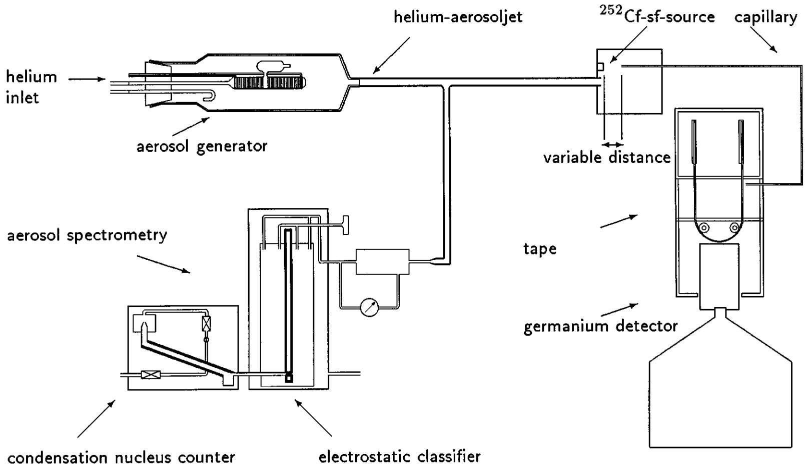

Fig. 2. Experiment at the ISN: Transport of radioactive ions on aerosols. Fission products of a ^{252}Cf-sf source are transported by the helium aerosol jet along a capillary to a tape and by the tape to a germanium detector in offline position. Parallel to γ-spectrometry, the aerosol production is surveyed by aerosol spectrometry using a negligible part of the volume

For the measurements of aerosols (size and concentration), a Differential Mobility Particle Sizer (DMPS) or a Scanning Mobility Particle Sizer (SMPS) and a condensation nucleus counter [9] are installed. The DMPS / SMPS measures the size distribution of submicrometer aerosols (10 nm to 1 μm aerodynamic diameter) by an electrical mobility detection technique. The particles are classified with an Electrostatic Classifier (Model 3071), and their concentration is measured with a Condensation Particle Counter (Model 3022) [10]. The experiment has the following free parameters:

1. the distribution of aerosols, variable in size and concentration;
2. the distance between source and extraction capillary (5 mm to 90 mm);
3. the collection time of aerosols on the tape;
4. the measuring time of the γ-spectra.

In addition to several fixed parameters (e. g. length of capillary, distance between sample and detector), these free parameters can be used to survey the mechanism of adsorption and the transport of fission products on aerosols. For example, aerosol size distribution characterized by mean diameter, total surface and concentration can be correlated to results obtained by γ-spectrometry: It is possible to increase for example the aerosol concentration while the aerosol diameter is kept fixed. On the other hand, the aerosol concentration can be held constant while the diameter is shifted.

The experiment shown in Fig. 2 was successfully installed in a test run at ISN. The measurement of the distribution of aerosols in the range of 10 nm to 1000 nm is performed every 120 s. The flow which is needed for this aerosol size spectrometry is only (3.0 ± 3.5) % of the total flow. The γ-spectrometry is applied for energies from 10 keV to 4000 keV. During this run, methods for selective collection concerning the half-life (choosing suitable collection and measuring times) were used and data concerning the correlation of aerosol distribution and collection efficiency of fission products on aerosols have been recorded.

3 Data analysis

The analysis of data is divided into two parts according to aerosol size spectrometry and γ-spectrometry. At the end of each analysis, the results are correlated. Data concerning the transport of radioactive ions on aerosols as a function of aerosol size distribution and element can thus be obtained. Each size spectrum of aerosols is analyzed individually. As an example, a spectrum with low aerosol concentration is given in Fig. 3.

When aerosols are used as a medium for the transport of radioactive isotopes, the stability of the aerosol distributions in concentration and size are essential for well-defined conditions during experiments. Moreover, the possibility of shifting the aerosol distribution to smaller or larger diameters (by increasing or decreasing the temperature of the wax vapour, s. Fig. 4) offers the opportunity of studying the mechanism and the selectivity of adsorption and transport of heavy ions on aerosols.

The γ-spectra are measured after an appropriate collection time of aerosols on the tape and a measuring time

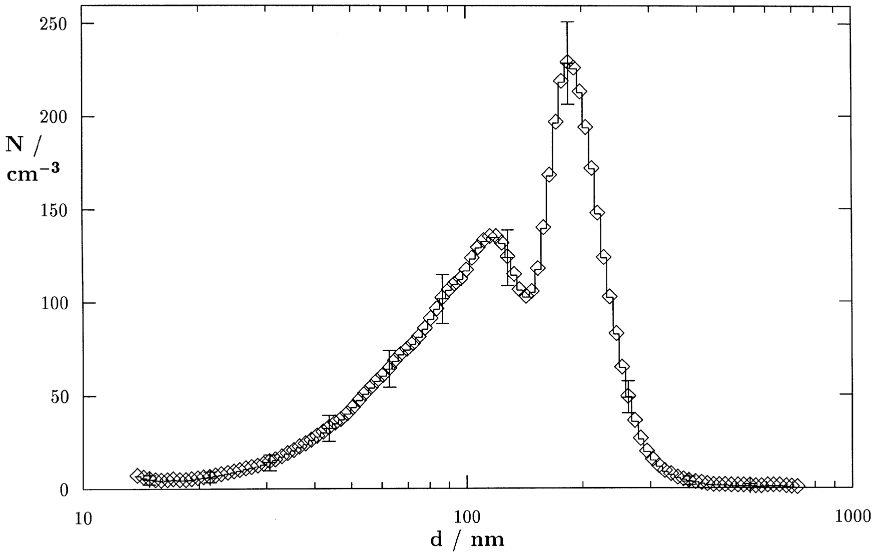

Fig. 3. Example of an aerosol spectrum measured with SMPS. The aerosols are produced by the aerosol generator (see Fig. 1) in a helium atmosphere and heated to a wax temperature of 485 ± 3 K. The figure shows the measured aerosol concentration N versus the aerodynamic diameter d (◇). Errors in the measurement of the concentration are marked by errorbars (representatively given every 10th step), refering to errors of measurement, statistics and calculations, while the intervals of detected aerodynamic diameters (which are equidistant on a logarithmic scale) are given by a stepping function

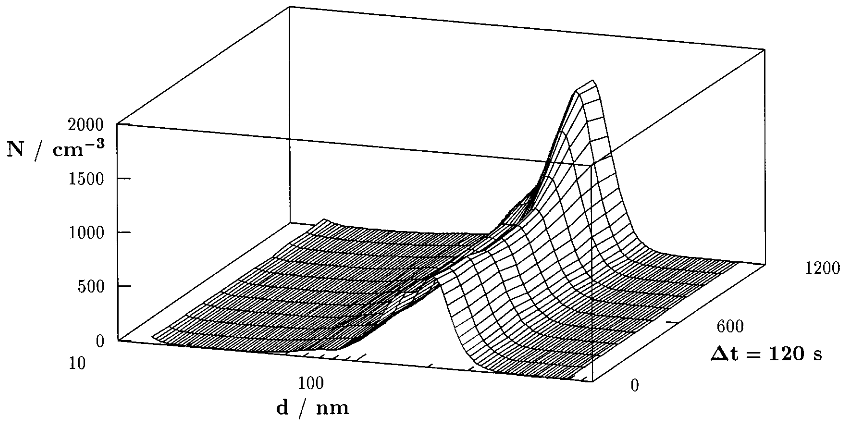

Fig. 4. Isometrical plot of aerosol spectra removed every 120 seconds. The figure shows the measured aerosol concentration N versus the aerodynamical diameter. Beginning at a stable distribution (t=0 s) at a wax temperature of 455 ± 3 K, another stable aerosol distribution (t=1200 s) is produced by increasing the temperature to 462 ± 3 K . The diameter of the critical nuclei is shifted from $d_* = 148.4 \pm 5.4$ nm to $d_* = 138.1 \pm 5.1$ nm by changing of temperature in compliance with the theory of the critical nucleus [11], and the concentration does rise about a factor of 2.4. Finally, the maximum of the distribution is moved from $d_{max} = 212.8 \pm 8.1$ nm to $d_{max} = 191.1 \pm 7.0$ nm

of the γ-spectra have been chosen, for example by setting of a half-life window. If, for example, a collecting time of 800 s is selected, while the measuring time for a series of γ-spectra is 400 s, the spectra measured during the first 400 s will yield the more short-lived isotopes and the second 400 s the longer-lived isotopes (Fig. 5). The comparisment of the last two peaks in this picture lead for example to an estimation of a half live of about (570 ± 200) s respectively (140 ± 80) s. Together with the energy and intensity of the peaks, this furnishes information allowing the nuclides to be identified: in this example ^{105}Tc and ^{109}Rh.

4 Transport of radioactive ions on aerosols - experimental results

By correlation of the results derived from γ- and aerosol size spectrometry, the efficiency of the ion transport as regards their aerosol parameters (concentration, radius, surface and volume) and the ions themselves (element and isotope) is obtained in this experiment for the first time. A comparison of measured and calculated adsorption rates of ions on aerosols is possible by a model of diffusion charging [8].

The measurement with a pure helium jet furnishes a count rate (transported ions identified by γ-spectrometry) which is compared to that of a helium aerosol jet as a function of the aerosol concentration. With the exception of the aerosol parameters, both measurements, pure helium jet and helium aerosol jet, are identical in geometry and other experimental conditions. Therefore, a direct comparison (e. g. ratio of counts) is justified. The ratio of counts for a special element, in this example Technetium (γ-rays of different Tc-isotopes with the mass numbers A = 101-106, with the exception of A=102), are calculated and given in Fig. 6 as a function of the energy E_γ for two fixed aerosol concentrations ((a): $6.0 \cdot 10^3$ cm^{-3}, (b): $4.5 \cdot 10^4$ cm^{-3}) relative to the measurements with a pure helium jet (ratio normalized to 1).

A first result is obtained by analyzing all elements from $_{35}$Br to $_{50}$Pr in the following manner: The rate of transported radioactive ions is element selective, and is not selective in isotopes. Furthermore, the addition of aerosols to the helium jet increases the transport efficiency of the jet at least by a factor of ten.

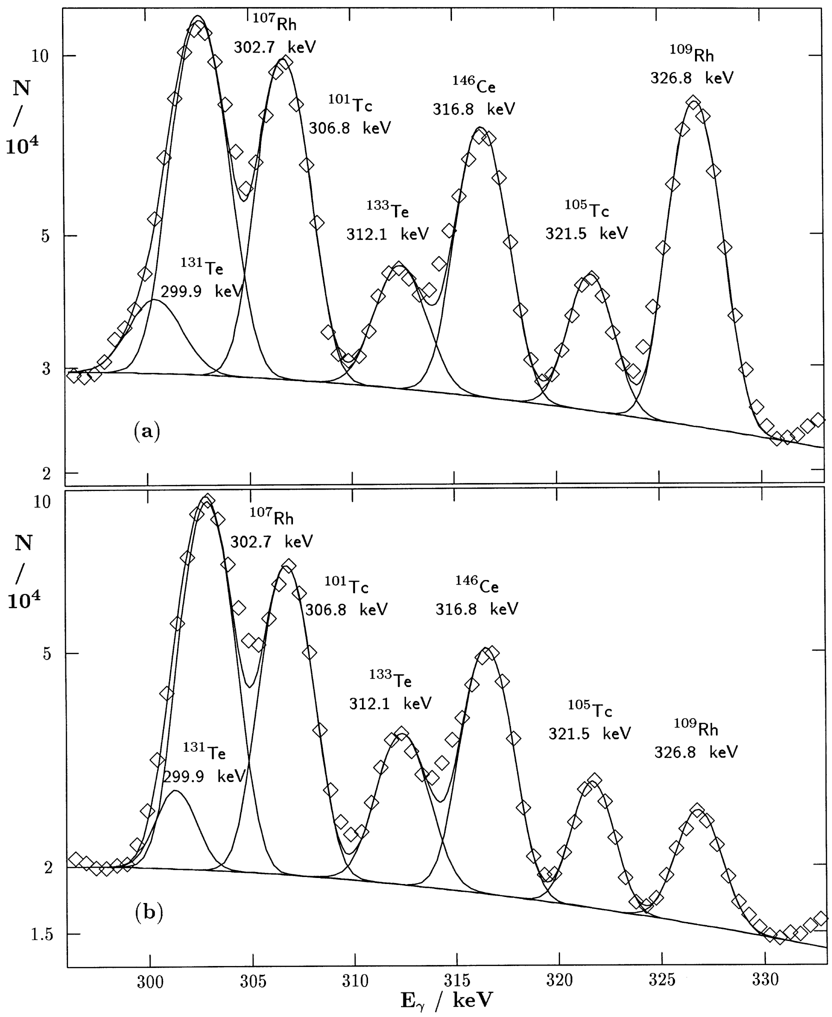

Fig. 5. a, b. Example of the analysis of a low energy part (296 - 333 keV) of γ-spectra summed up. Each γ-spectrum (in total 2 $*$ 51 spectra) is measured during a time of 400 s, while the collecting time is 800 s. Thus the first summed spectrum (a) has a delay time of t_d = 0 s while that of the second spectrum (b) has one of 400 s

The Technetium isotopes for example, shown in Fig. 6 yield that even the low aerosol concentration (a) increases the rate of counts by about a factor of 14.8 ± 1.1, while the high aerosol concentration (b) results in a factor of 51.6 ± 3.4. When another two aerosol concentrations $2.0 \cdot 10^4$ cm^{-3} and $2.9 \cdot 10^4$ cm^{-3} are taken into account, a saturation function of transported ions versus aerosol concentration can be calculated. The best type of fitted function for this saturation curve is an exponential:

$$\frac{Z_{He+A}}{Z_{He}} = a \cdot exp(-\frac{b}{N_a + N_0}) \quad \text{with} \quad N_0 = \frac{b}{ln\ a} \qquad (1)$$

Z_{He+A}: counting rate with helium aerosol jet, Z_{He}: counting rate with helium jet, N_0: constant for normalization, b: inclination coefficient, a: limiting value in case of the total transport, N_a: aerosol concentration.

In (1), the inclination of the exponential function is characterized by the coefficient b while the constant a gives the limiting value for the ratio of counts in the case of an aerosol concentration $N_a \to \infty$. In practise a transport of 95% will be sufficient in most cases, which results to a desired aerosol concentration of $5.7 \cdot 10^4$ cm^{-3} for Bromine and of $3.8 \cdot 10^5$ cm^{-3} for Palladium (the concentration of other elements is between these values) based on the activity of the ^{252}Cf-sf source. Up to now, the aerosol generator has been capable of producing an aerosol jet of 10^8 cm^{-3}, the required concentrations are therefore easily established.

The data is also analyzed for the dependency of the transport of radioactive ions on aerosols as a function of the aerosol radius or surface and volume.

Based on a model for diffusion charging, a linear dependency relating the number of ions as a function of the aerosol radius is expected for the adsorption process [8].

The calculation model is rather simple but efficient. The adsorption of ions is dominated by the electric charge and results in a state of equilibrium of the diffusion charging [12] and the electric field on the aerosol [13]. Assuming that an aerosol is at rest in its environment, the transport of ions is governed only by the diffusion equation modified by the electrical forces arising from the free charge carried by the aerosol.

In the experiment, this adsorption is measured using a source with an activity of 7.0 MBq. For low aerosol con-

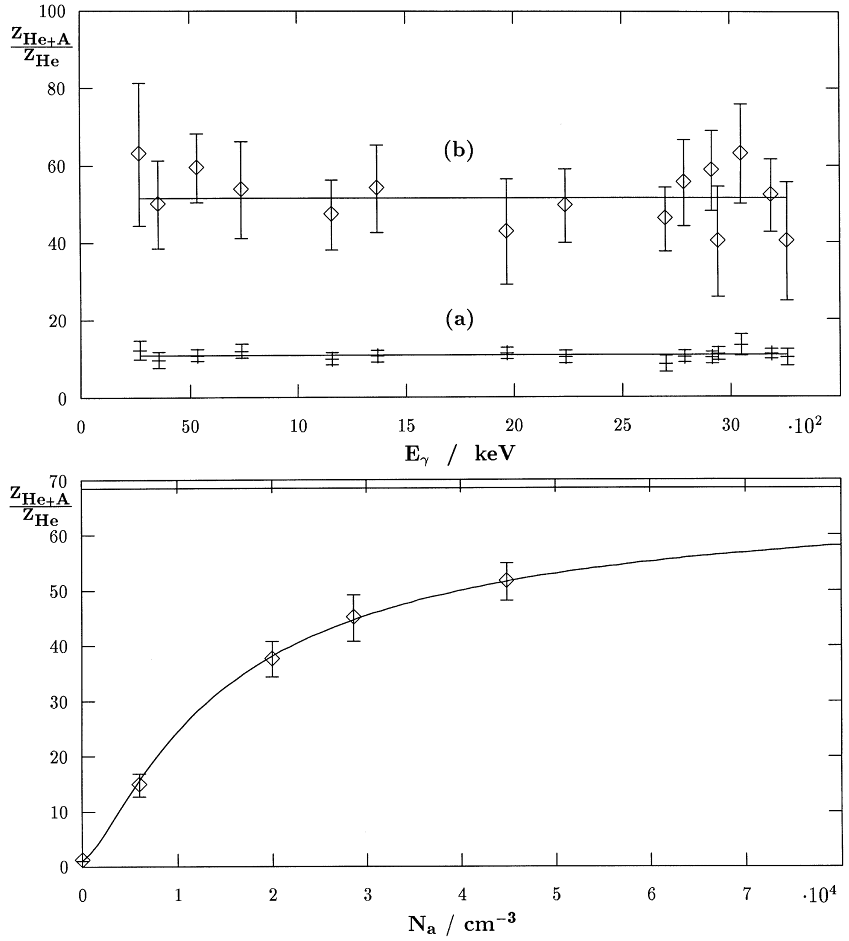

Fig. 6. a, b. Rate of counts for different Technetium isotopes measured with a helium aerosol jet relative to a pure helium jet as a function of the energy and the aerosol concentration. The results of the γ-spectra (a) correspond to an aerosol concentration of 6.0 $\cdot\ 10^3$ cm^{-3} while those of (b) belong to a concentration of 4.5 $\cdot\ 10^4$ cm^{-3}. Independent of isotope or energy, the rate of transported ions is improved by a factor of 14.8 ± 1,1 (a) and 51.6 ± 3.4 (b). Two other aerosol concentrations, 2.0 $\cdot\ 10^4$ cm^{-3} and 2.9 $\cdot\ 10^4$ cm^{-3}, are taken into accout for the saturation function in the figure below. Saturation means transport of every ion produced and results in a calculated factor of 68 ± 4

centrations, the predicted linearity is measured, for higher aerosol concentrations, saturation is observed. This implies that, for high aerosol concentrations, all ions produced can be collected, see Fig. 7.

Linear regression yields:

$$\frac{Z_{He+A}}{Z_{He}} = 1 + (29 \pm 3) \cdot 10^2 \mathrm{m}^{-1} \cdot \frac{O}{d_m} \qquad (2)$$

with a correlation coefficient of $R^2 = 0.99$

Z_{He+A}: counting rate with helium aerosol jet, Z_{He}: counting rate with helium jet, O: total surface and d_m: mean diameter.

When data for different elements (from Bromine to Palladium) are combined and the results for the transport efficiency analyzed (see example in Fig. 6), the maximum rate of transported ions can be correlated with the element normal potential N$_p$, see Fig. 8.

Linear regression yields:

$$N_I = (72.2 \pm 0.8) - (2.2 \pm 0.4) \cdot N_p \qquad (3)$$

with a regression coefficient of $R = 0.75$.

N_I: rate of collected ions and N_p: element normal potential.

These extensive evaluations of the experimental data yield the following principal results:

1. Adsorption and transport of ions on carnauba-wax aerosols are element selective.
2. Adsorption and transport of ions on carnauba-wax aerosols increase linearly with the aerosol radius.
3. Carnauba-wax aerosols carry a well-defined number of ions in the equilibrium state (for example r = 100 nm: 26 ± 9 ions, on an average for the multi-ion source ^{252}Cf-sf).
4. A highly concentrated aerosol jet is capable of collecting all ions produced.

5 Possible adaptions and conclusions

Using carnauba-wax for the generation of aerosols, helium aerosol jet turned out to be the most effective medium for the transport of radioactive ions. It is at least ten times more effective than a pure helium jet. The aerosols produced are spherical, and variable distributions can be achieved, with the mean diameter ranging from 10 nm to 800 nm. The concentration of aerosols can be regulated independently, from 10^3 cm^{-3} to 10^8 cm^{-3}. A highly concentrated helium aerosol jet is capable of transporting all

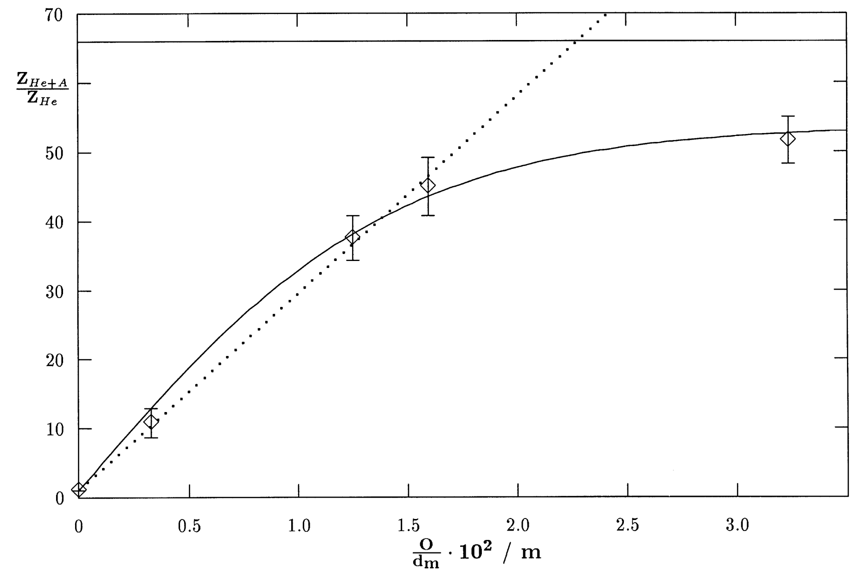

Fig. 7. Rate of collected ions as a function of the total aerosol surface per mean diameter. For this measurement, different Technetium isotopes are analyzed. The deviation of the linear dependence is observed for aerosol concentrations higher than $2 \cdot 10^4$ cm^{-3} and results in a saturation curve

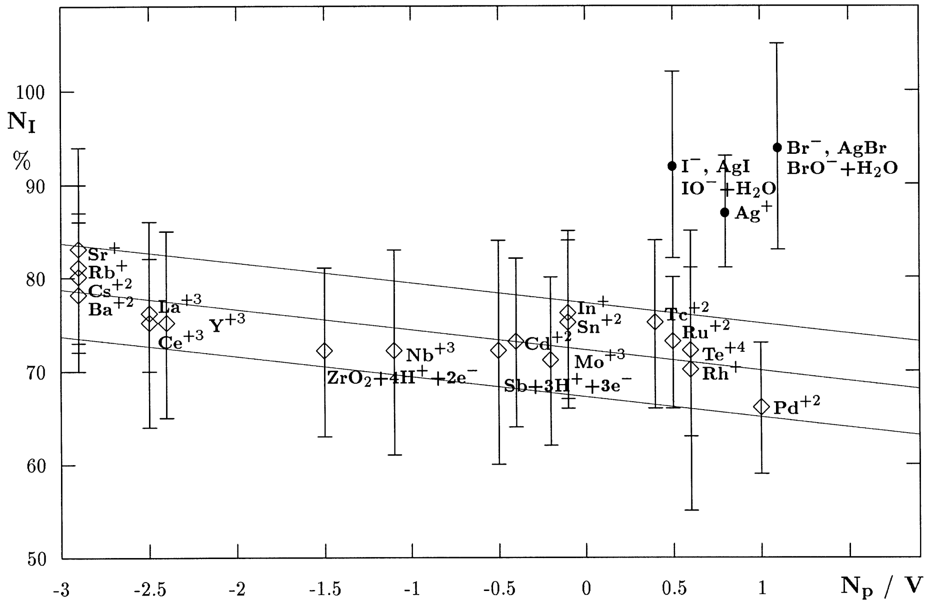

Fig. 8. Maximum rate of collected ions N_I for an aerosol concentration of only $4.5 \cdot 10^4$ cm^{-3} as a function the element normal potential N_p of metal ions (◇). In some cases, typical chemical reaction channels are listed for further clarification. Halogenes and silver (●) are excluded

ions produced. This new aerosol generator is therefore a valuable tool for the fast transport of short-lived exotic isotopes, for example.

The method of carrying out aerosol size spectrometry and γ-spectrometry in parallel provides information about the transporting flow, fast, on-line and with high resolution in the aerosol size spectra.

The adsorption and transport of radioactive ions on carnauba-wax aerosols is element selective and not selective in isotopes. It is possible to correlate the collection efficiency of each element with its normal potential and thus optimize the required aerosol jet in compliance with the needs of the experiment. Furthermore, the collection efficiency rises linearly with the aerosol radius (outside the region of saturation), which agrees remarkably well with model calculations of diffusion charging of aerosols.

As to the problem of collecting short-lived isotopes at accelerators, mass separators or Radon reference chambers, the new aerosol generator offers several possibilities. The transporting flow is mixed with well-defined carnauba-wax aerosols, either in constant distribution or monodisperse size. Ions are adsorbed to aerosols and transported along the flow. The aerosols are not only used as the transporting medium; the experiment will also yield data about the aerosols themselves, for example charge as a function of size and selectivity of adsorption in order to study most effective collection of exotic ions. Combining the aerosol collection techniques, the tape system for example, with on- or offline $\beta\gamma$-coincidence measurements [14], the study of very neutron-rich isotopes is possible due to the very high collection efficiency and the very large solid angle.

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