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IEEE Transactions on Nuclear Science, Vol. NS-32, No. 5, October 1985

BEAM DIAGNOSTICS BY X-RAY SPECTROSCOPY AT THE UNILAC

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## Abstract

At the Unilac a set-up for element analysis of the beam, based on x-ray spectroscopy, was tested. The ions are excited by a carbon foil, the element composition of the beam is determined from the spectra of the emitted radiation. Elements with an atomic number of 15 or greater can be detected in concentrations down to  $0.1^{\circ}_{\circ}$ .

# Introduction

The Unilac is used to accelerate a great variety of elements from oxygen to uranium. Particular attention must be given to the injector section to deliver all isotopes without impurities. Identification and separation of the nuclides is done by a magnetic spectrometer which resolves differences in the charge to mass ratio of less than 0.4%. But this is not sufficient to exclude all possible contributions of unwanted elements: if the charge to mass ratio differs only slightly or is the same like  ${}^{96}Mo^{4+}$  and  ${}^{1*4}Sm^{6+}$  the isotopes cannot be separated.

To analyse the contents of the beam a set-up based on x-ray spectroscopy was tested. The ions are excited by a carbon foil and the emitted radiation is analysed by a semiconductor detector. Like in microprobe analysis the element composition of the beam can be determined from the spectra: the energy of the lines give the atomic number Z, their intensity the proportion in the beam.

### Apparatus

The beam energy must be high enough to excite sufficient radiation even for high atomic numbers. So the set-up was installed in the stripper section at a beam energy of 1.4 MeV/u. A schematic view of the apparatus is shown in fig. 1.



Fig. 1: Schematic view of the apparatus.

As target material carbon was chosen for its good thermal stability and its low energy characteristic radiation which is not seen by the detector. Therefore no target lines occur in the spectra. Different foils with area densities of 30 and 100  $\mu$ g/cm<sup>2</sup> can be moved into the beam.

. The x-rays are observed outside the vacuum by a 10  $\,\rm mm^2~Si(Li)$  - detector, mounted perpendicular to the

beam to reduce the doppler shift. The detector had to be shielded from the high level of x-radiation in the accelerator tunnel which causes a detector count rate of 350 cps. This could be reduced to about 0.4 cps by surrounding the detector with a lead shield of 4 cm thickness.

The window in the vacuum chamber is made by a 50  $\mu$ m Hostaphan foil whose absorption of radiation is acceptable. For further reduction of low energy radiation (e.g. the intense M-series) aluminium foils with thicknesses from 10 to 250  $\mu$ m can be placed in front of the detector. The total rate of x-ray photons can be varied continuously by attenuation of the Unilac beam.

During operation of the accelerator access to the apparatus is not possible. Thus target foils and aluminium absorbers are remotely controlled. To maintain the detector permanently cooled an automatic liquid nitrogen refilling system had to be installed.

Data acquisition is done by a multichannel analyser which has built-in functions for energy calibration and determination of peak net area, centroid and FWHM. It is linked to a personal computer to store and plot the spectra.

### Element identification

The atomic number is determined from the energy of the spectral lines. These energies differ from the well tabulated values for neutral atoms, the lines are shifted to high energies due to the quite high ionization of the beam [1]. The shift decreases with increasing Z, it is up to about 100 eV for transitions between the inner shells and up to more than 1 keV for transitions involving outer shells. Values measured during the tests are plotted in fig. 2.



Fig. 2: Observed shifts of characteristic x-ray lines.

Very low energy radiation is not seen by the detector due to absorption in the Hostaphan window and in the air between window and detector which gives a lower limit of the photon energy of about 2 keV. This restricts the detectable elements to Z greater or equal 15 (phosphorus).

## Quantitative determination

The number  $N_{\underline{X}}(\lambda)$  of photons of energy  $E_{\underline{\lambda}}$  from a transition  $\lambda$  emitted when  $N_p$  particles impinge on a thin target of area density  $n_{T}$  is given by

$$N_{\chi}(\lambda) = \sigma(\lambda) n_{T} N_{P}$$
(1)

where  ${}_{\zeta}\sigma(\lambda)$  is the x-ray production cross section. From these photons only a small portion is seen by the detector due to the limited solid angle  $\Omega$ , the detector efficiency  $\epsilon(E_{\chi})$  and attenuation by a factor  $A(E_{\chi})$  caused by matter in the photon path. Thus the number of counts  $N_{C}(\lambda)$  in the spectrum is

$$N_{C}(\lambda) = N_{X}(\lambda) \Omega \varepsilon(E_{\lambda}) / A(E_{\lambda}) .$$
 (2)

The attenuation  $A(E_{\lambda})$  depends on thickness d and attenuation coefficient  $\mu(E_{\lambda})$  of the absorbing material. If the radiation has to pass a layer of different materials with d<sub>i</sub> and  $\mu_i(E_{\lambda})$  then  $A(E_{\lambda})$  is given by

$$A(E_{\lambda}) = e^{\sum \mu_{i}(E_{\lambda}) d_{i}} .$$
(3)

According to (1) - (3) the number of particles  $N_p$  can be determined from the number of counts  $N_{\mbox{C}}(\lambda)$  of a spectral line. But for element analysis only relative intensities are required so it is sufficient to determine the ratio of the number of particles  $N_p^1$  and  $N_p^2$  of two elements in the beam from the number of counts of two of their spectral lines  $\lambda_1$  and  $\lambda_2$  which is given by

$$\frac{N_{P}^{1}}{N_{P}^{2}} = \frac{N_{C}(\lambda_{1}) A(E_{\lambda_{1}})}{N_{C}(\lambda_{2}) A(E_{\lambda_{2}})} \frac{\sigma(\lambda_{2}) \varepsilon(E_{\lambda_{2}})}{\sigma(\lambda_{1}) \varepsilon(E_{\lambda_{1}})} .$$
(4)

Here  $\Omega$  and  $n_T$  cancel and thus only  $\sigma(\lambda)$ ,  $A(E_{\lambda})$  and  $\epsilon(E_{\lambda})$  must be taken into account.

The efficiency  $\varepsilon$  was measured using calibrated radioactive sources [2], it is 1.0 in the energy range from 5 to 15 keV and drops to 0.8 at 2 keV and 0.4 at 30 keV. The x-ray attenuation by the Hostaphan window, the aluminium absorbers and the air between window and detector is calculated from tabulated values of  $\mu_i$  [3],

it is plotted in fig. 3.



Fig. 3: Attenuation A of x-radiation.

The cross sections  $\sigma(\lambda)$  are shown in fig. 4 for excitation of K- and L-series in collisions with carbon at an energy of 1.4 MeV/u. The dots represent data from the literature [4] where all except the Pb-L cross section are for carbon projectiles. The crosses are measured with the described set-up according to (1) - (3) where  $N_{\rm p}$  is determined using standard Unilac beam instrumentation. The solid lines show the result of the BEA-theory [5]. All data are in sufficient agreement.



Fig. 4: Cross sections for the production of projectile K and L radiation in collisions with carbon at an energy of 1.4 MeV/u; •: data from [4], +: determined with element analysis set-up, solid line: BEA theory [5].

Due to its simplicity the K-series are favoured for element identification. But from fig. 4 it is seen that the cross section decreases rapidly with increasing Z. Thus for high atomic numbers the excitation of the K-series is to weak and for Z greater than about 50 the L-series has to be used for identification.

# Performance

#### Counting time

The Unilac delivers beam only during short macropulses (repetition rate 50 Hz) with a dutycycle of 25% or less. To avoid pulse pile-up, the count rate of the detector has to be kept low enough during the macropulse which results in an upper limit of the mean count rate of 100 cps. With this rate 5 to 10 minutes are needed to get sufficiently clear spectra.

# Resolution

The energy differences of spectral lines from adjacent elements in the periodic system are big enough to determine the atomic number of the main content in the beam, so an injector setting to a wrong element will be detected easily.

The detection of contaminations is complicated by the limited resolution of the detector: the lines of small contributions may be hidden by low intensity lines of the main element. To get information about the detection limit in such cases a test was made using a calcium ion source sputtered with argon to produce a mixture of  ${}^{*0}Ca$  and  ${}^{*0}Ar$  in the beam where the intensity ratio of the components was changed by variing the source parameters.

The difference of the energies of the argon  $K_\beta$  and the calcium  $K_\alpha$  line is only 230 eV whilst the FWHM of these lines is 270 and 190 eV so they cannot be fully separated. The observed spectra are shown in fig. 5. It is seen that the calcium contribution of 4% leads only to a small change in the line shape of the Ar $K_\beta$  line.

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These small deviations can be detected without difficulty if the spectrum of the main element is known. To ease the interpretation of measured spectra a catalogue of reference spectra is in preparation.



Fig. 5: Spectra of a mixture of calcium and argon. Upper plot: 20% Ca, 80% Ar; lower plot: 4% Ca, 96% Ar. Dotted line: spectrum of pure argon, fitted to Ar K<sub>a</sub> line. REC: radiative electron capture.

If the lines of impurities and main content do not overlap they can be detected if their intensity is higher than the statistical fluctuations of the background in the spectra. As an example in fig. 6 the detection limit of contaminations in an argon beam is shown, calculated from a typical argon spectrum (counting time 10 min). It is shown the portion of contaminations for which the number of counts of the  $K_{\alpha}$ respectivly the L line is higher than the square root of the number of background counts, taken within the FWTM of this line. The detection limit increases for increasing atomic number Z since the x-ray production cross section and also the detector efficiency for K radiation for Z > 40 decrease. The poor values for medium and high Z are not as critical as in fig. 5: the low energy Ar K-radiation may be suppressed by the aluminium foils, enhancing the sensitivity for the high energy radiation of high  $\bar{Z}$ elements. For elements other than argon the results are



similar.

Fig. 6: Detection limit of contaminations in terms of main element intensity for an argon beam.

# Example

As an example where contaminations were detected the spectrum of a  $^{76}$ Ge beam taken during routine operation of the Unilac is shown in fig. 7. Besides the Ge K lines also Ar and Fe lines are seen, indicating unexpected portions of 15% Ar and 0.05% Fe in the beam. It turned out that the argon peak is from the rare isotope  $^{36}$ Ar (abundance only 0.063%) of the argon auxiliary source-gas, the iron peak may result from excitation of the stainless steel walls of the beam tube by scattered ions but a small lineshift to high energies indicates that it more likely results from a contribution of  $^{57}$ Fe (abundance 2.2%) from mechanical components in the source. For injection into the prestripper  $^{76}$ Ge<sup>4+</sup> was chosen, so  $^{38}$ Ar<sup>2+</sup> and  $^{57}$ Fe<sup>3+</sup> had the same charge to mass ratio and could not be separated in the injector.



Fig. 7: Spectrum of a <sup>76</sup>Ge beam, taken during routine operation of the Unilac. Argon radiation partly suppressed by 10 μ aluminium. REC: radiative electron capture.

## Acknowledgement

The author would like to thank the members of the atomic physics group at GSI, especially D. Liesen, for their valuable advices and R. Schwedhelm and his group for their support in installing the setup.

# References

- F. Folkmann, "Progress in the description of ion induced x-ray production, theory and implication for analysis", in <u>Ion beam surface layer analysis</u>, vol. 2, O.Meyer, G.Linker and F.Käppeler ed., New York: Plenum Press, 1976, pp. 695-718.
- [2] D.H. Hoffmann; private communication.
- [3] <u>Handbook of Spectroscopy</u>, vol. 1, J.W. Robinson ed., Cleveland: CRC Press, 1975, ch. III, pp.28-154.
   [4] R.M. Wheeler et al, "K x-ray production cross sec-
- [4] R.M. Wheeler et al, "K x-ray production cross sections for fourteen elements from calcium to palladium for incident carbon ions", <u>Phys. Rev. A</u>, vol. 13, pp. 958-964, March 1976.
  T.J. Gray et al, "K-shell x-ray production cross sections for <sup>12</sup>C, <sup>14</sup>N, and <sup>16</sup>O ions on Ni, Rb, Ag, and Sb: 0.4-2.4 MeV/amu", <u>Phys. Rev. A</u>, vol. 13,

pp. 1344-1351, April 1976. W.A. Schönfeldt, thesis, GSI Report 81-7, Darmstadt, West Germany, July 1981.

[5] J.D. Garcia, R.J. Fortner, T.M. Kavanagh; "Inner-Shell Vacancy Production in Ion-Atom Collisions", <u>Rev. Mod. Phys.</u>, vol. 45, pp. 111-177, April 1973.