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DEVELOPMENT OF A SCINTILLATION
DETECTOR FOR FAST ION COUNTING
IN A MASS SPECTROMETER

by

H. A. TASMAN

1970



Joint Nuclear Research Center
Karlsruhe Establishment - Germany
European Institute for Transuranium Elements

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ABSTRACT

A description is given of a scintillation detector, designed for fast counting of 3 kV heavy ions. It applies intermediate acceleration to the incident ions to permit reliable operation with 60 kV conversion voltage. The pulse height spectrum measured at the detector output allows the estimation of the counting efficiency as well over 95 % with a background of only 3 counts per second. A experimental correction formula for counting losses, based on a dead time of 25.2 ns, together with additional corrections at very high as well as at very low counting rates, seems to hold up to about 5 Mc/s. However, in spite of apparently correct performance, isotope ratios measured show a scatter of $\pm 1\%$, which is two to three times the scatter from a standard open multiplier detector. The origin of the scatter could not be traced. In the present state, the scintillation detector should not be preferred for measurement of ion beams of more than 10^{-15} A, since the standard multiplier offers a better reproducibility at lower complexity and cost.

KEYWORDS

SCINTILLATION COUNTERS
HEAVY IONS
MASS SPECTROMETERS
ACCELERATION
CONVERSION
ELECTRIC POTENTIAL
LOSSES
ISOTOPE RATIO
MEASUREMENT

Development of a Scintillation Detector for fast Ion Counting
in a Mass Spectrometer *)

General Remarks

All information carried by the ion beam in a mass spectrometer might be exploited to its fullest extent, if one had a means of counting the ions faithfully. This requires a detector with good time resolution and accurately predictable dead-time-losses, faithful response, and a good signal-to-noise ratio. At present, no commercially available detector is known to be fully satisfactory for the detection of 3 kV heavy ions. The standard detector for low intensity ion beams is an open electrostatically focused secondary electron multiplier. This detector works reasonably well if the anode current is integrated and recorded in analog form or digitized subsequently. When the anode current exceeds about $1 \mu\text{a}$, non-linear effects occur, which have a long time constant. The accuracy on small signals is limited through the statistical spread in the number of ions per time constant of the detecting system, as well as by amplifier and detector noise. Amplifier noise is usually negligible, the statistical spread can be accounted for by subsequent data processing. Detector noise is mainly present in "dirty" multipliers, and may be reduced by cleaning through high vacuum bake-out.

When anode current is integrated before it is processed, variations in multiplier gain affect the result directly. In isotope ratio measurements, only moderately short-time fluctuations interfere, occurring in a time comparable with the durations of one scan. Such deviations have been observed with anode currents above about $1 \mu\text{a}$. However, experience shows that reproducibilities of 0.3 - 0.5% may be obtained with an open multiplier and analog recording.

*) Manuscript received on 22 October 1970

For direct ion counting, the open multiplier is not particularly suited. With the first dynode at negative HT (about - 2 kV for a 17-stage multiplier), the ions hit it with about 5 keV energy (3 kV acceleration + the negative multiplier voltage). At this energy, the conversion factor at the first dynode η (= the mean number of electrons per impinging ion) is about 2 - 3. The counting losses due to ions not releasing any secondary electron from the first dynode, are roughly $\exp(-\eta)^2$. Any fluctuations in η affect the observed counting rate, they affect the integral multiplier gain in a more serious manner, however.

A further limitation of the usefulness of the available 17-stage multiplier ^{†)} lies in the fact, that it cannot be operated over appreciable periods of time at a gain of more than about 1×10^6 , as this causes a rapid decrease in performance. The best 100 Mc/s pulse preamplifier that was available for the present investigation, allowed the discriminator threshold to be set equivalent to an input charge of 10^6 electrons per pulse, thus requiring an integral current gain of substantially more than 10^6 . Operation at a gain of 10^7 appeared to reduce the multiplier life to 3 weeks.

It should be noted, however, that satisfactory ion counting might also be possible with an open multiplier, in which the number of stages has been increased to furnish a sufficient gain without excessive gain per stage. It remains to be investigated how much of the deterioration at high gain is due to excessive output currents, which remain unaffected by distributing the required gain over a larger number of stages.

^{†)} 17-Stage open multiplier in Atlas- CH-4 mass spectrometer.

The main justification for using a scintillation detector for ion counting is founded on the more favorable pulse height distribution at its output, when compared with a standard open multiplier detector. This difference is primarily due to the higher conversion ratio from ions to secondary electrons, η , as a consequence of the higher ion energy on impact on the conversion electrode.

Scintillation Detector (Daly-Type)¹⁾

Principle of Operation

After passing through the grounded collector defining slit, the positive ions are deflected sideways onto a conversion electrode at a high negative potential. The resulting secondary electrons are accelerated onto a scintillator at ground potential, which is incorporated in the wall of the vacuum chamber. The scintillations generated by these electrons are detected by a commercial photomultiplier outside the vacuum. The design corresponds roughly to Fig. 1, without items 7, 8, 9, 10, 13, 15 and 16.

This arrangement offers the advantage, that the ions impinge on the conversion electrode with a considerably higher energy than on the first dynode of an open multiplier. With a conversion voltage of -50 to -60 kV, the mean number of electrons per heavy ion of 3 keV energy, η , is about 8. The number of photoelectrons from the photomultiplier cathode is of the order of 100 per incident ion, so that essentially no further counting losses occur, once at least one secondary electron is ejected from the conversion electrode. The photomultiplier dark current can easily be discriminated. Contrary to the open multiplier, the gain of the sealed photomultiplier is unaffected by organic contamination of the detector. The commercial photomultiplier can be exchanged easily, without breaking the vacuum.

The (plastic) scintillator is aluminized on the side facing the vacuum for three reasons :

a. It is essential that a conducting layer be present to eliminate the charge of the impinging electrons,

b. The aluminium coating nearly doubles the gain, by reflecting the light, that goes in the wrong direction, back onto the photomultiplier,

c. The aluminium coating reduces the vaporization of the scintillator into the vacuum.

For very low counting rates only, consideration c. could be circumvented by using a glass scintillator, which offers the additional advantage of being bakeable at high temperature, whereas the plastic scintillator softens at 80°C . However, only fast plastic scintillators offer sufficiently short decay times to allow counting rates over 1 Mc/s with acceptable dead time corrections.

The aluminium coating should be thick enough to reflect blue light essentially completely, yet not thicker than necessary, such as to limit electron energy loss in the aluminium. The optimum is said to be around 500 \AA . This gives about 80 % reflection at 400 nm and 5-10 keV energy loss.³⁾

Modification of the Daly design to detect 3 keV ions.

The Daly design¹⁾ consists of a negative conversion electrode, off-axis in an otherwise grounded environment. A high η is obtained using oblique incidence and an impact energy of about 40 keV.

A similar arrangement was built. It worked well with Xe^+ ions of at least 8 keV energy, applying - 30 kV to the conversion electrode. When the geometry is to be retained, such as to retain oblique incidence, lowering the ion energy necessitates a proportional reduction of the conversion voltage. With 3 keV ions and -11 kV conversion voltage, η will be reduced to a value close to what is obtain-

ed in an open multiplier. The gain of the assembly is decreased far more drastically, however, as the aluminium coating on the scintillator absorbs a fixed amount of energy of about 5-10 keV. Under these conditions, serious interference was observed from the photomultiplier dark current.

A higher conversion voltage could possibly be retained, when the conversion electrode would be retracted further away from the unperturbed ion trajectory. This possibility has not been tested.

As a more elegant solution, an arrangement was realized and tested, in which the conversion electrode was surrounded by a cup at an intermediate potential. This arrangement provides a forward acceleration to the ions on entering the cup. It reduces the field emission from the conversion electrode by reducing the field strength. Finally, the entry aperture acts as a two-element converging lens. A schematic representation of the modified detector is given in Fig. 1.

Experimental Apparatus

Mass Spectrometer

The detector was built for a CH-4 Mass Spectrometer (Atlas Werke, Bremen/Germany), which is a 60° sector instrument with the ion beam in a vertical plane. Its performance was tested with Xe^+ -ions produced by electron impact, as well as with Re^+ , W^+ , and U^+ -ions from thermal ionization. In all cases, the ion acceleration voltage was 3 kV. Although the detector output could also be integrated, and displayed through the standard DC-amplifier on an analog strip chart recorder, most of the testing was done by counting the ions directly. Details of the counting and data collection system are given below.

High voltage supplies

The negative conversion voltage of up to -60 kV was taken from a Brandenburg Model 906 supply (Brandenburg Ltd, South Croydon, Surrey, England). The stability of $\pm 0.4\%$ as well as the current of 0.4 mA turned out to be amply sufficient. The intermediate acceleration voltage was supplied by a 10 kV, reversible polarity, high voltage unit, made by the Zentralwerkstatt GmbH, Göttingen/Germany, this unit, available from a previous project, was certainly overdimensioned, as the current demand was very small, and variations of several % could be tolerated without any effect on the counting efficiency.

High voltage for the photomultiplier was taken from the standard multiplier supply in the CH-4 mass spectrometer.

Constructional details of the Detector

General

Fig. 1 shows a schematic representation of the detector, on which all essential elements are drawn to scale.

The conversion electrode (11) was made from Duraluminium (AlMg3), highly polished by mechanical means, the final polish being done with a household metal polish paste. Cup (9), cover (10), scintillator cup (19), as well as the housing and flanges, were made from stainless steel, electrolytically polished. Cup, cover, and scintillator cup were given a final mechanical polish with Nr. 400 emery cloth. The insulators 12, 13, and 16 are high purity alumina.

The conductance of the tubular connection through 7 to the mass spectrometer analyzer is so small, that a separate pumping system is indispensable. A 15 l/s Vac-Ion ion-getter-pump is connected to the housing through an optically dense baffle, blackened internally with gold black (Fig. 4). The scintillator (18) is sealed to the

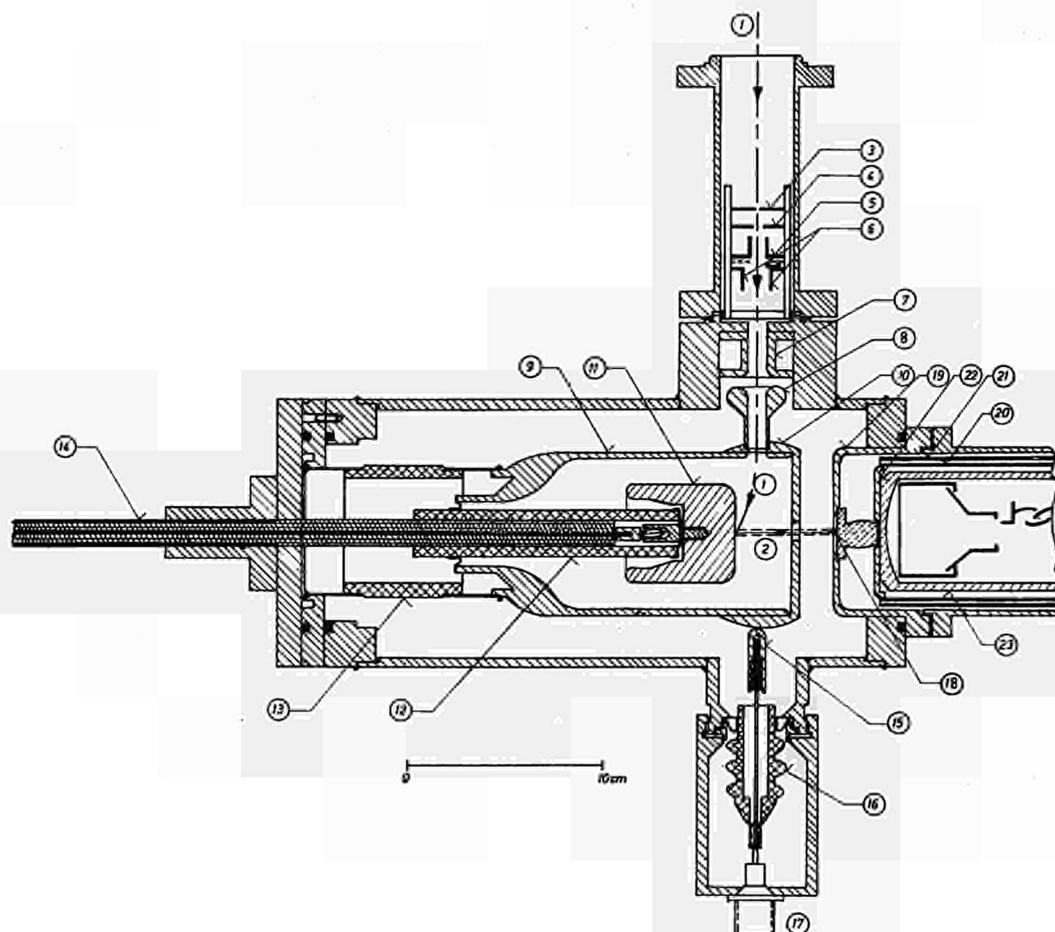


Fig. 1 Slightly schematized cross-section of the modified scintillation detector.

1 : incident ion beam, 2 : secondary electron beam, 3 : collector defining slit, 4 : secondary electron suppressor for Faraday cup collector, 5 : Faraday cup collector, with retractable bottom, 6 : beam centering plates, 7 - 8 : two element tube lens, 9 : cup at intermediate - EHT, 10 : cover, 11 : conversion electrode, 12 : 60 kV alumina insulator; vacuum tight, 14 : EHT cable, coaxial, 15 : spring loaded cup contact, 16 : 15 kV alumina insulator, vacuum tight, 17 : intermediate - EHT connector, 18 : aluminized scintillator foil on light guide, 19 : scintillator cup (at ground potential), 20 : inner magnetic shielding at photocathode potential (negative HT), 21 : outer magnetic shielding at ground potential, 22 : Armco disc for magnetic shielding of the photocathode, 23 : photomultiplier (56 AVP or 56 DVP-03)

cup (19) with an indium wire gasket, the scintillator cup (19), the 10 kV insulator (19), the 10 kV insulator (16), as well as the flange on which the insulator (13) with the cup (9) is mounted, are sealed with Viton gaskets.

Conversion electrode

Several conversion electrode shaped were tested (Fig. 2 and 3). The original shape (Fig. 2 A) showed a sharp increase in background counting rate when the conversion voltage was made to exceed - 40 kV. This was thought to be due to luminous pre-breakdown discharges along the insulator surface. The shape of Fig. 2B, chosen such as to minimize surface charge build-up at the insulator near the cathode, gave a marked improvement. The best results were obtained with the shape of Fig. 2 C. Advancing the conversion electrode (Fig. 2 D) blurred the secondary electron image on the scintillator.

Attempts to focus the image by means of a concave conversion electrode were only moderately successful. Tests were made with concave spherical radii of 15, 22, and 30 mm (Fig. 3). 15 mm gave overfocusing, 30 mm under-focusing, whereas with 22 mm the spot position on the scintillator remained stationary, when, with -60 kV conversion voltage, the cup voltage was varied from -6.5 to -9 kV. Due to mechanical asymmetry of stray magnetic fields, however, the spot was not centered on the scintillator. Moreover, when a minor breakdown occurred with the concave 22 mm electrode, - 60 kV conversion voltage being applied through a 100 MegOhm series resistor, the focused secondary electrons destroyed the scintillator dramatically, with a flat face electrode (Fig. 3C) a similar event did not destroy the scintillator.

Ion-Getter-Pump Interference

A 15 l/s Vac-Ion pump, branched directly to the detector

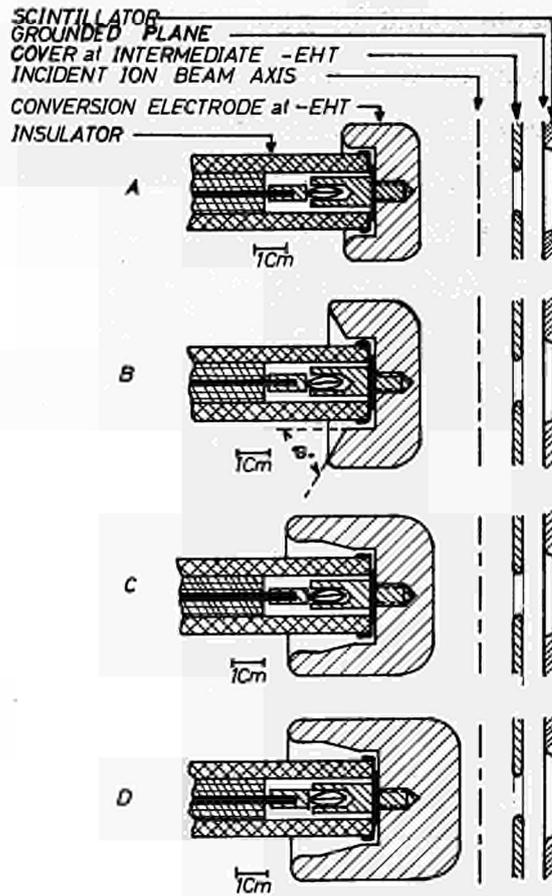


Fig. 2. Conversion electrode shapes.
The best results were obtained with the shape c, which was retained in the final version.

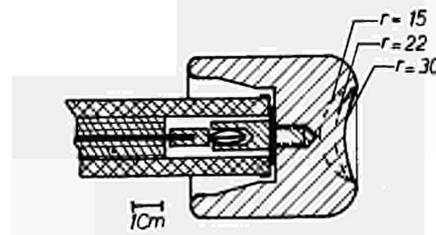


Fig. 3. Hollow conversion electrodes focused the secondary electron beam, but presented additional hazard for the scintillator. Optimum focusing was obtained with 22 mm radius.

housing, injected a background of about 10^5 c/s at a pressure of 2×10^{-7} Torr. A grounded fine mesh grid, mounted in the pump aperture had essentially no effect at all. A right angle bend or valve reduced this interference only by a factor 10. An optically dense baffle gave a reduction by a factor 500. Blackening the interior surfaces with gold black (Fig. 4) (gold evaporated in an atmosphere of 1 Torr Argon) raised the attenuation factor to 10^5 , which was considered sufficient, as the usual operating pressure is well under 10^{-7} Torr.

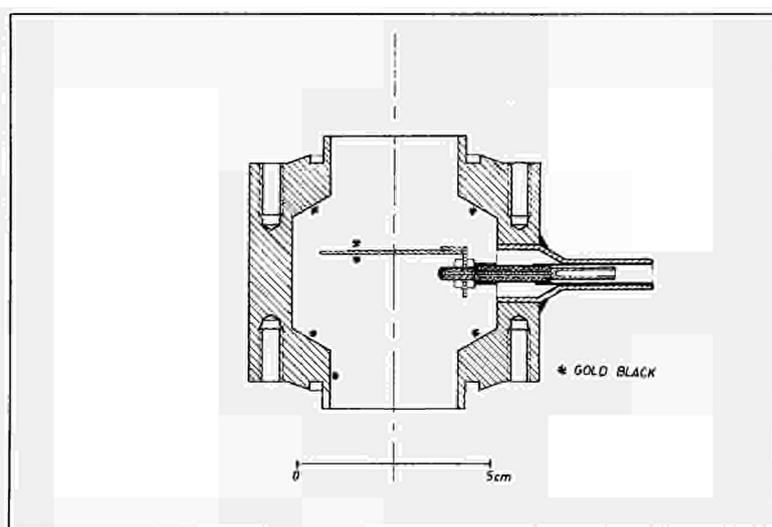


Fig. 4. Baffle for the suppression of ion-getter-pump interference. The inner surfaces (*) are blackened with gold black. Interference attenuation factor ca. 10^5 .

The interference should not be caused by charged particles from the pump discharge, as it is unaffected by a high negative or positive potential on the baffle disc. It is not caused by light quanta from the pump, that reach the photomultiplier directly, as it is only seen when a high negative conversion voltage is applied. It is thought to be due either to UV quanta, or to excited neutrals, that eject electrons on impact with the conversion electrode.

Vacuum

After a mild bake-out (40-50°C) a (pump) pressure of 2×10^{-8} Torr is obtained, notwithstanding the presence of the (aluminized) plastic scintillator.

Scintillator and Light Guide

Two makes of commercial fast plastic scintillators have been used, Naton 136 (Thorn Electronics Ltd), and NE-104 (Nuclear Enterprises Ltd), both as solid material, and as thin foil of 0.2-0.3 mm thickness.

The solid material turned out to be difficult to polish, it showed a "smeary" behavior (more so than plexiglass which polishes quite well), and should present the disadvantage of being much thicker than the penetration depth of 60 kV electrons (< 0.1 mm). However with such a gain/discriminator setting, that the photomultiplier dark pulses were not detected, neither the solid material nor the 0.2-0.3 mm foil did contribute to the background. We used the foil, which was cemented on a plexiglass disc with light guide (Fig. 5). None of the cementing techniques tried was completely satisfactory. The best results were obtained with NE-580 two-component optical cement (Nuclear Enterprises Ltd). A viscous solution of plexiglass in chloroform, or Eastman Cement Nr. 910, both gave less satisfactory results : initially the joints appeared correct, but after a few days small cracks and/or milky clouds developed. Reasonable joints were also obtained with Canada Balsam, which did not set sufficiently, however.

The plexiglass light guide of toroidal shape approximates the ideal logarithmic spiral, and is much easier to realize. Attempts to make the light guide from polyvinyltoluene (the solvent of the plastic scintillator) failed, as it was impossible to polish it well enough because of its "smeary" behavior.

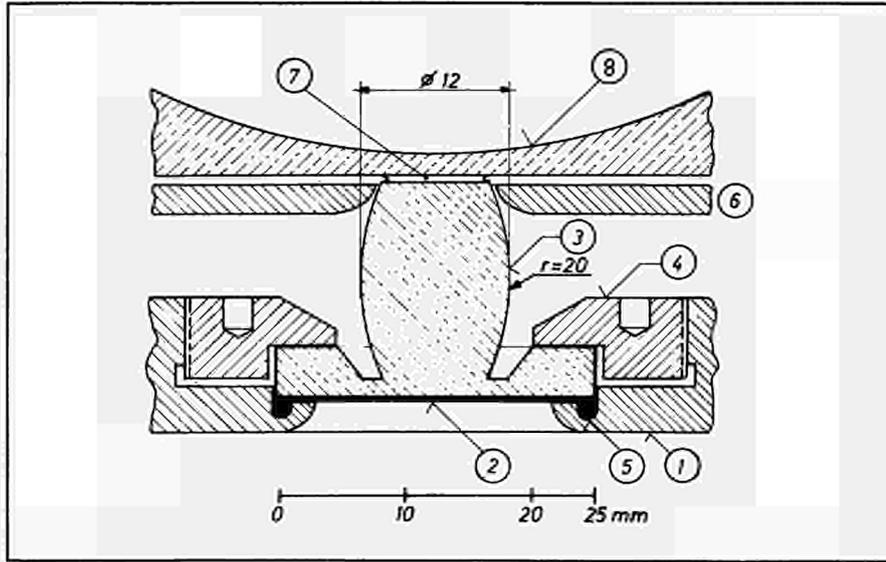


Fig. 5 : Scintillator and light guide

- 1 : scintillator cup, 2 : aluminized scintillator foil,
3 : perspex light guide, 4 : threaded retaining ring, 5 : indium wire seal,
6 : Armco disc for magnetic shielding of the photocathode,
7 : optical coupling liquid, 8 : photocathode of photomultiplier.

with
The scintillator was coated a 500 \AA aluminium layer in a vacuum of 2×10^{-5} Torr or better. At first, the thickness of the coating was controlled by "weighing" it with a quartz crystal mounted aside the substrate. Then it was found, however, that a coating with a reflection and transparence apparently equivalent to that of a weighed layer of $500 \text{ \AA} \pm 20 \%$ could be obtained by a quick total evaporation of a weighed amount of aluminium from a flat tungsten boat straight under the substrate, assuming a cosine distribution to determine the amount of aluminium to be flashed.

In early experiments with Naton 136, a slow decay was observed of the light emission after a strong ion beam was switched off abruptly. An apparently exponential decay occurred with a time constant (reduction by a factor e), that depended on the multiplier gain

and discriminator setting, and that was as long as 1.2 seconds for a gain such that the multiplier dark pulses were just eliminated. At lower gain, the apparent time constant decreased, amounting to 30 ms under normal operating conditions. Visual inspection showed this "afterglow" to be greenish-yellow, in contrast to the blue light of the scintillations.

Subsequent experiments with NE-104 did not show this phenomenon, an upper limit for any afterglow decay constant could be set at about 1 ms. However, in one of the later experiments, a discharge occurred, which partially damaged the thin aluminium coating on the scintillator. Subsequently, a slow decay with an apparent time constant of 0.2 s was also observed in NE-104. Consequently, at least in this case, the afterglow phenomenon seems to be related to slow elimination of the charge accumulated during the strong excitation, rather than to an inherent radiation transfer property of the scintillator itself.

Photomultiplier and pulse shaping

The photomultiplier 56AVP (Radiotechnique/Valvo) proved to be satisfactory. A somewhat better signal-to-noise ratio at high gain settings was obtained with the 56DVP-03 of the same manufacturer.

The pulse duration at the multiplier output was too long to exploit fully the available fast electronics. It was found possible to reduce this pulse duration by means of a passive filter ⁵⁾ of 50 Ohm iterative impedance, located in the multiplier housing. The filter reduces the charge per pulse by about a factor 4, which can readily be compensated by an increase in multiplier gain. It proved to be important to use low inductance carbon resistors for the filter as well as for the final stages of the multiplier bleeder chain. The improvement obtained is shown in Figs. 6 and 7., Fig. 8 shows the filter, Fig. 9 the bleeder. For these measurements, the scintillator was excited

directly with a β -emitter (Tl-204), such as to minimize the spread in pulse shape in view of the detection with a sampling oscilloscope.

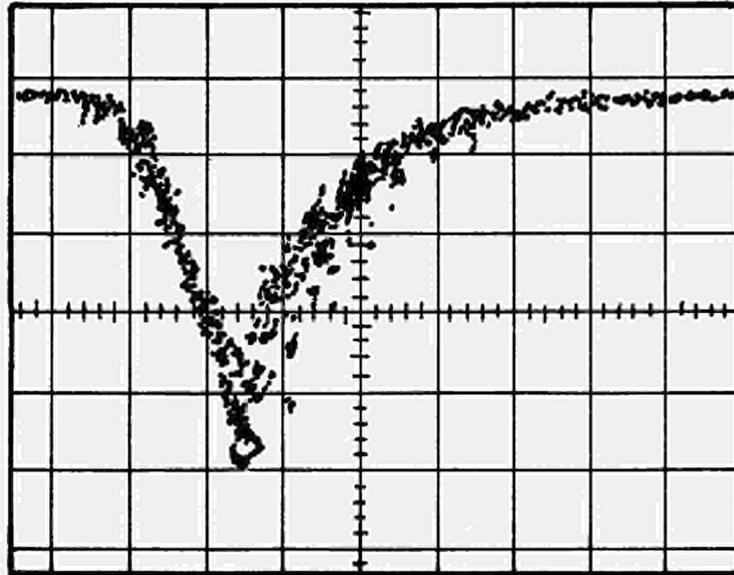


Fig. 6. Output pulse at the photomultiplier anode, without passive filter, scintillator in mounting of Fig. 5 excited with β -radiation.

Scintillator : NE-104

Photomultiplier : 56 AVP

Excitation : Tl-204

Horizontal : 4 ns/cm

Vertical : 100 mV/cm (50 Ohm)

Photomultiplier voltage : -2016 V on bleeder chain of Fig. 9.

The light guide concentrates the light on a circular spot of 10 mm diameter. It should be possible to reduce the effective photocathode area by making the focusing electrode Foc in the multiplier negative with respect to the cathode K. However, when doing so, no increase in signal-to-noise ratio was observed experimentally, not even at gain settings so high, that the multiplier dark pulses were counted.

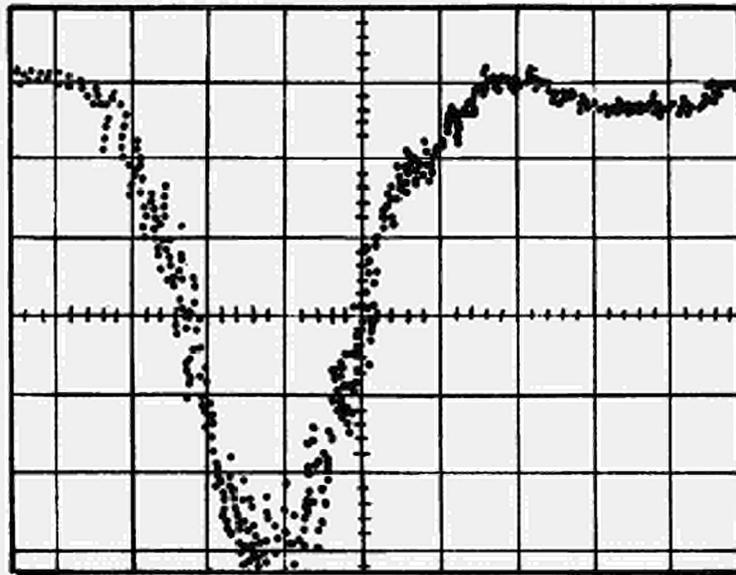


Fig. 7. Output pulse at photomultiplier anode, with passive filter (Fig. 8), obtained under similar conditions as Fig. 6.

Scintillator : NE-104

Photomultiplier : 56 AVP

Excitation : T1-204

Horizontal : 2 ns/cm

Vertical : 100 mV/cm (50 Ohm)

Photomultiplier voltage : -2083 V on bleeder chain of Fig. 9.

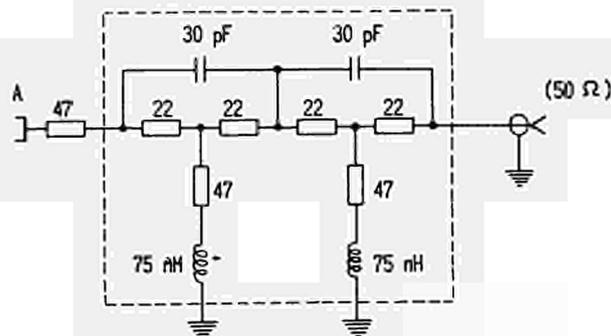


Fig. 8. Passive filter for pulse shortening.

All resistors are 1/4 Watt low inductance type.

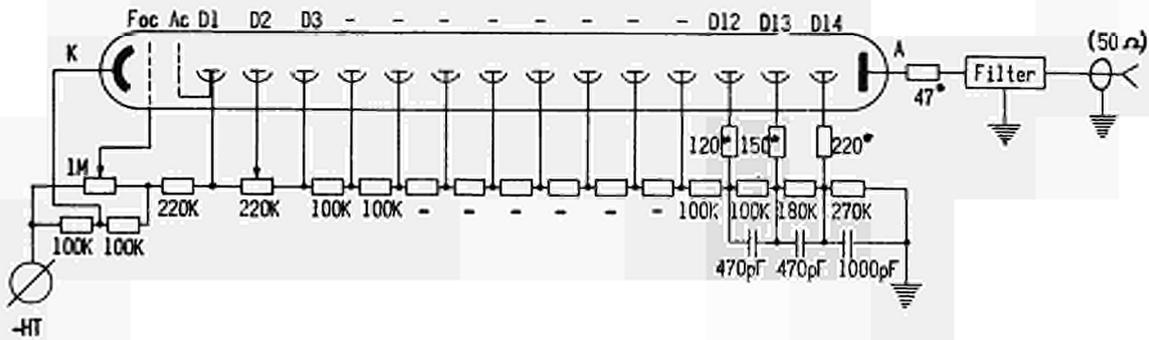


Fig. 9. Bleeder chain for the photomultiplier 56 AVP (56 DVP-03). The resistances marked with an* are low inductance type 1/4 Watt.

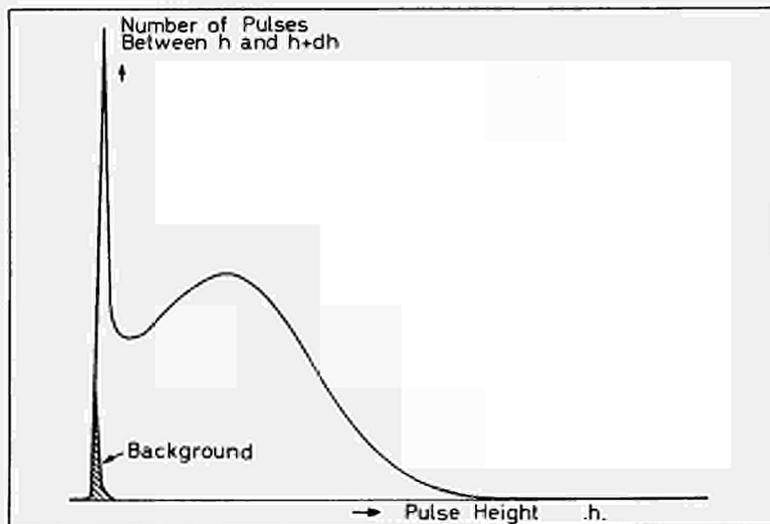


Fig. 10. Pulse height spectrum from scintillation detector. Excitation : 3 kV Xe⁺ ions, conversion voltage :-50 kV. Units arbitrary.

Pulse height distribution

The more favourable pulse height distribution of the scintillation detector was verified experimentally with a multichannel-analyzer. Even without the filter of Fig. 8, the pulse duration was

too short for correct operation, and had to be lengthened by a pulse stretcher. The counting rate had to be limited to very low values. The pulse height spectrum obtained is shown in Fig. 10. It is to be compared with Fig. 11, which shows the spectrum obtained under similar conditions from a standard open secondary electron multiplier. As both spectra are represented in arbitrary units, only the general appearance of the curves is to be compared. It should be noted, that the pulse stretcher and/or the pulse height analyzer did not perform completely satisfactorily, in particular, it was not possible to locate the zero on the pulse height axis accurately, and the large sharp peak near zero pulse height might be due to instrumental defects. The shaded part of it, named "background", was recorded during a subsequent counting period of equal length, with the ion beam suppressed but all other conditions remaining unchanged.

From Fig. 10 it can be estimated, that the discriminator and gain can be set such as to eliminate virtually all background, whilst retaining a counting efficiency of well over 95 %. Unfortunately, a slight instability in the mass spectrometer source control did not allow a direct measurement of the counting efficiency, which could be determined by comparing the observed counting rate from a stable beam, with the integrated current of a Faraday cup ahead of the scintillation detector ((5) in Fig. 1).

Fast electronics

The pulses from the filter output are fed into a fast charge sensitive amplifier, followed by a discriminator with fixed threshold and a scale of 10, all of which are contained in a small metal box, fitted directly to the multiplier housing. This unit (Zchn. Nr. 42306-19) was designed specially for this purpose by Frieseke + Hoepffner GmbH, Erlangen-Bruck/Germany. The equivalent input sensitivity is 10^6 electron charges, the time resolution, tested with a pulse generator, is better than 10 ns. The amplifier comprises a zero level restoration circuit to avoid pile-up, the equivalent discriminator

stability, referred to the input, should be better than $\pm 2\%$. The scale of 10, which is neither reset nor read-out, results in a large decrease in bandwidth required in the subsequent counter, and furnishes a digital signal level, high enough to make the unit insensitive to disturbances such as switching transients. A Tesla discharge (Leybold High Frequency Vacuum Tester) can be sprayed directly on the amplifier box without any adverse effects.

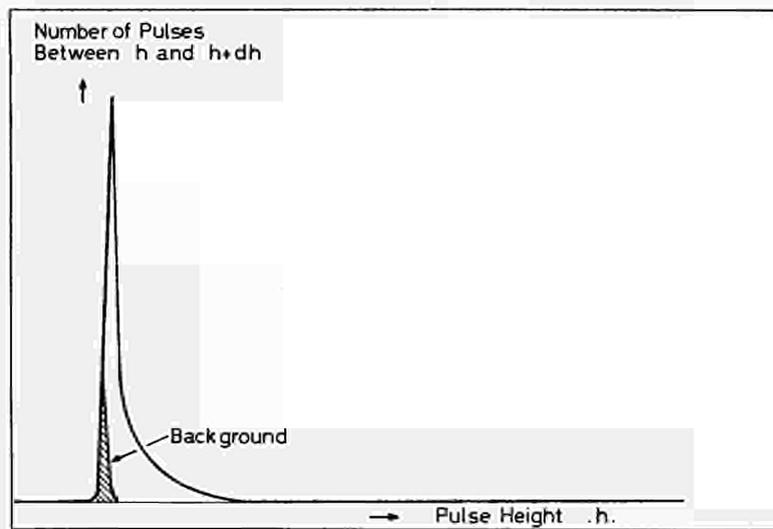


Fig. 11. Pulse height spectrum from standard secondary emission multiplier. Excitation : 3 kV Xe^+ -ions, First dynode at - 2 kV. Units arbitrary.

Data collection

For data collection, part of a system was used, that was developed in the Labor für Elektronik, Gesellschaft für Kernforschung, Karlsruhe ⁶⁾. A short description follows belows (See Fig. 12) :

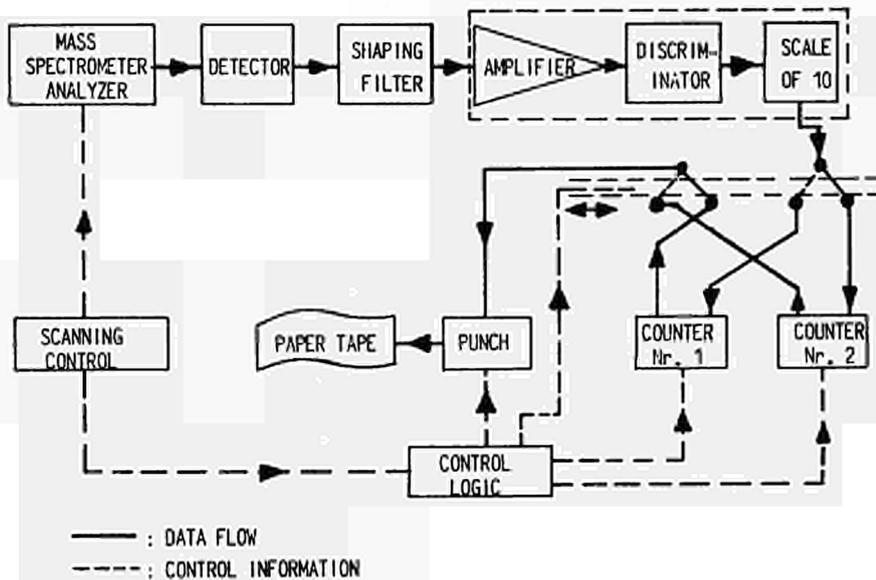


Fig. 12. Schematic representation of the data collection system.

A control logic directs the output of the scale of 10 alternatively to either one of two counters; whilst counting occurs in one counter, the contents of the other are read out and punched in paper tape, after which that counter is reset. The punch is a Tally, Model 420, 75 char./sec. The present logic punches 4 characters of address and control information plus 6 decimal characters per counting interval. Counting is started after a start pulse derived from the magnet scanning control of the mass spectrometer, after which a fixed number of 400 points or intervals are counted. The punch limits the minimum counting time per point to 0.3 seconds, which setting was used in all experiments. A simple mechanical timing control starts the scanning repetitively at 128 s. intervals.

The present control unit had been designed as a prototype. Unfortunately, it is wasteful in tape (producing 10 meters per scan), and lacks flexibility because of the limited speed and the fixed number of points per scan.

Data Reduction

A computer program (DATA 3 - DATA 4) was written for

the IBM-1130 digital computer. This program is an adaptation of the DATA 1 - DATA 2 program ⁷⁾, which digests mass spectra recorded on paper tape through a "dataprint" digital voltmeter (Varian MAT, Bremen/Germany).

An analysis or run consists of a series of up to 12 successive scans. On the output tape, a separation between that last scan of a run and the first scan of the next run, is made by manual intervention. In short, the program performs the following operations:

1. The program can accept up to 8 successive runs on one continuous tape. It requests at first the "label date", consisting of the number of runs, date, analysis numbers, element names, masses, writes these in a file on the disc, and retrieves them as necessary for the processing of the subsequent runs.

2. The peaks are assumed to be flat-topped, the top slope must not necessarily be zero. The flat top range is not constant, but depends on the resolution and the source settings. The program accepts a single slow scan, preferably of one peak, to extract a peak shape. This calibration should be specified accordingly by requesting only one mass. Straight lines are fitted iteratively through the base line and top regions. The peak shape, defined as the top slope times the half width divided by the peak height, as well as the range of the flat top, are recorded on the disc for later reference.

3. The tape is read scan by scan, checking for code errors. In the first scan of each run, the highest peaks are searched, until the number requested in the label has been found. The peak positions are checked against the specified masses for mass scale consistency. Through each flat top range, a straight line is fitted iteratively with the slope imposed by the latest peak shape calibration. In subsequent scans, peaks are only searched in the regions around the previously found peak positions.

4. The peak heights are linearly interpolated to the starting point of each scan, from the second scan onward, to correct for ion current decay. The isotope ratios are computed, applying "mass discrimination" correction factors, retrieved from a file on the disc, for the element and masses in question. The isotope ratios are checked for outliers, applying the Dixon outlier criterion.

5. The printed output consists of a heading with the "label data", followed by the mean isotope ratios, and isotope percentages (atomic and by weight), together with the corresponding standard deviations computed from the spread of the results from the successive scans.

Results of Isotope Ratio Measurements

1. Xenon.

Xenon was selected, because all interfering background peaks could be reduced to a negligible level by bake-out of the ion source region and analyzer at 200-300°C. The mass spectrum was recorded at several different inlet pressures, whilst retaining all other parameters constant. The computed isotope ratios, referred to mass 124, were divided by their values from the literature, and are plotted as a function of the observed counting rate in Fig. 13.

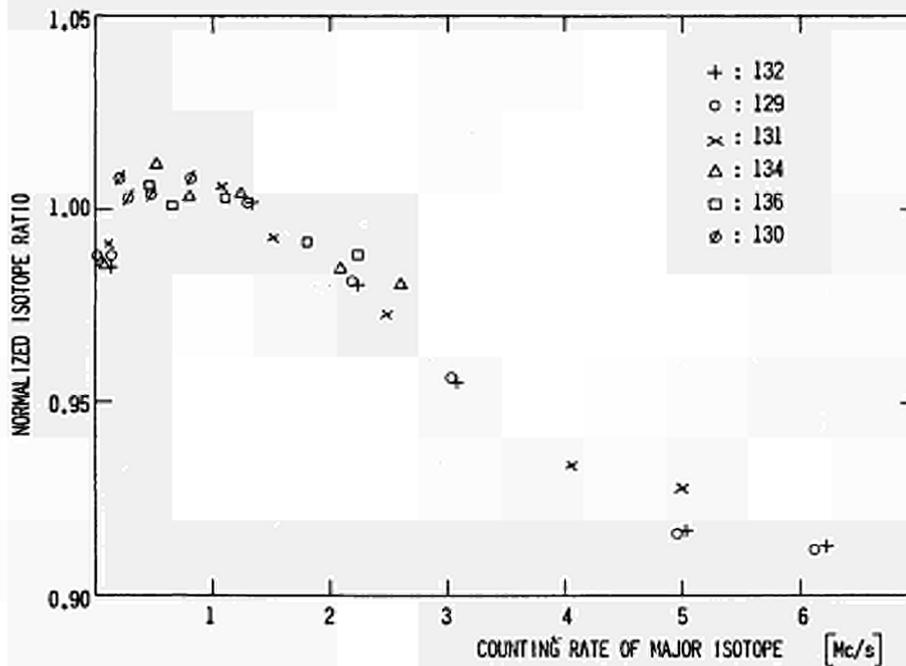


Fig. 13. Normalized Xe isotope ratios as a function of the counting rate of the major isotope. The abscissa shows the ratio of the abundance of the indicated isotope to that of Xe-124, divided by the corresponding ratio computed from literature⁹⁾ values.

This "normalized ratio" can be reduced to unity \pm 0.7% (Fig. 14), when the counts collected in the time t are corrected by the following (experimental) formula :

$$N_{\text{corr}} = N_{\text{obs}} \frac{1 + A \exp(-B N_{\text{obs}}/t)}{1 - N_{\text{obs}} \tau/t + C(N_{\text{obs}}/t)^6} \quad (1)$$

where :

N_{obs} , N_{corr} are the observed and corrected counts

t is the counting time in seconds

$$A = 0.04$$

$$B = 0.16 \text{ s.}$$

$$\tau = 25.2 \times 10^{-9}$$

$$C = 0.86 \times 10^{-36} \text{ s}^6$$

τ can be interpreted as a dead time. The meaning of the exponential

correction in the numerator is not clear. The sixth-power-correction with coefficient C might be due to imperfect base line stabilization in the preamplifier, together with imperfect matching, allowing reflections to be counted when the base line is shifted due to pile up at high counting rates.

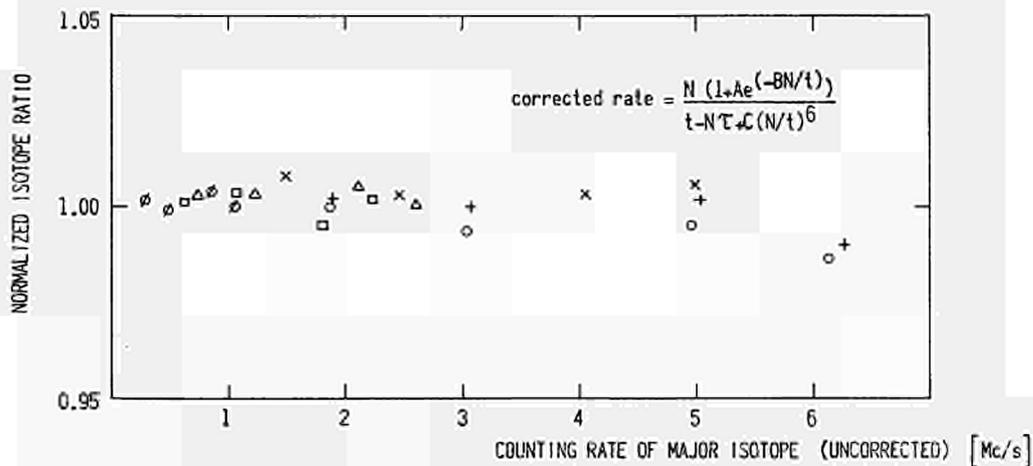


Fig. 14. Normalized Xe isotope ratios, represented in the same way as in Fig. 13, but applying the indicated correction formula to the observed counting rate.

N = number of ions counted during time t.

A = 0.04

B = 0.16 sec.

C = $0.86 \times 10^{-36} \text{ sec}^6$

= 25.2 nsec

2. Rhenium, Tungsten.

The mass spectrum, obtained through thermionic ionization of the filament material was recorded at different temperatures. The results, corrected through expression (1), are represented in Tables 1 and 2, in which NS is the number of scans, CR (i) is the corrected counting rate on peak i (mean value) (in Mc/s), and the error limits are the standard deviations from the spread of the ratios from the successive scans, referring to the single

measurement rather than the mean.

3. Natural Uranium.

A sample of natural uranium (about 1 μg), was analyzed with a two-filament cartridge. The results are shown in Table 3. The counting rates have also been corrected through formula (1).

Discussion.

Despite a low background counting rate and an empirical "dead time" correction formula (1) that seems to hold up to 5 Mc/s, a scatter of the results of about $\pm 1\%$ persists. This is also reflected in the standard deviations of the single measurements, derived from the results from successive scans. The measured 187/185 ratio in rhenium is about 2% lower than the literature value, this discrepancy is certainly larger than any correction for "mass discrimination" derived from integrated ion beam measurements. The low values for the 180/183 ratios in tungsten, as well as the high 184/183 ratio of the last tungsten run, remain unexplained.

For ion currents exceeding about 10^{-15} A or 6 kc/s, the scatter from the present scintillation detector counting system is definitely worse than the reproducibility obtained with a good standard electron multiplier, the output of which is integrated before further processing. The origin of the scatter could not be traced definitely. Part of the scatter could arise from instability in the mass spectrometer itself.

On the other hand, the last uranium run demonstrates the potential usefulness of ion counting for the measurement of very small ion currents. The intensity of the U-235 beam was only 3×10^{-17} A or 200 c/s. The scan speed was such that the top of the peak was measured during about 3.3 seconds per scan. Consequently, a statistical spread of 4% could be expected, which is not exceeded in

the actual experiment.

Comparison of scintillation detector versus open multiplier with
with subsequent digitization

The alternative open multiplier system that is to be compared with the scintillation detector system as used in the present investigation, is supposed to consist of a housing with an open multiplier, the output of which is digitized with a voltage to frequency converter. The other items, such as the collector slit system, the negative high voltage for the multiplier, and the data collection system (counters, control logic, output unit such as tape punch) are supposed to be identical for both alternatives, and do not enter in the comparison.

1. Equipment cost

<u>Scintillation Detector</u>		<u>Open Multiplier</u>	
Detector system	\$ 2500	Housing	\$ 250
Photomultiplier	\$ 150	Open Multiplier	\$ 625
- 60 kV supply	\$ 750		
- 10 kV supply	\$ 250		
100 Mc/s Electronics (Pre-amplifier, Discriminator, Scale-of-10)	\$ 1250	V/F Converter	\$ 750
	<hr/>		<hr/>
	\$ 4900		\$ 1625

2. Reliability

The additional units required for the scintillation detector system represent possible sources of trouble. When assembling the detector system, extreme care must be taken to assure cleanliness and absence of dust, more so than with the open multiplier. Minor contamination may be cleaned by a gas discharge in about 1 Torr Helium or Argon ⁸⁾. The open multiplier, on the other hand, may

Table 1
Rhenium Isotope Ratios

187/185	NS	CR(187)
1.663 \pm 0.023	6	0.41
1.653 \pm 0.023	5	0.43
1.662 \pm 0.003	5	0.23
1.659 \pm 0.013	4	6.3
1.698	Literature 9)	

Table 2
Tungsten Isotope Ratios

180/183	182/183	184/183	186/183	NS	CR(184)
0.00814 \pm 0.00021	1.848 \pm 0.018	2.137 \pm 0.020	1.959 \pm 0.030	7	0.23
0.00833 \pm 0.00013	1.855 \pm 0.023	2.130 \pm 0.025	1.956 \pm 0.028	9	1.00
0.00846 \pm 0.00038	1.828 \pm 0.025	2.226 \pm 0.030	1.985 \pm 0.080	6	1.15
0.00938	1.834	2.128	1.973	Literature 9)	

Table 3
Natural Uranium Isotope Ratios

235/238	NS	CR (238)
0.007197 \pm 0.000109	8	0.27
0.007121 \pm 0.000110	7	0.30
0.00746 \pm 0.000145	4	0.027
0.007258	Literature 9)	

be cleaned by baking at high temperatures, the removal of noise-provoking deposits on insulators not necessarily being as effective, as the signal path from multiplier to output does not contain a discriminator. When electrical breakdown is avoided, the scintillation detector is quite insensitive to overload from high ion currents. The life of the open multiplier is severely limited under these conditions. Overload of the photomultiplier results in an increase of background, part of which restores with time. Exchanging the photomultiplier is a simple procedure, without breaking the vacuum.

3. Reproducibility

The scintillation detector shows a scatter of the results of roughly $\pm 1\%$, which seems to be independent of the counting rate, from 400 c/s, up to 5 Mc/s. This compares unfavorably with the reproducibility of $\pm 0.3 - 0.5\%$, obtained with a standard open multiplier for all but the weakest ion beams. For ion beams exceeding 10^{-15} A or 6000 c/s, the standard multiplier is more accurate. For weaker beams, direct counting offers an advantage, although part of the scatter from the standard multiplier can be eliminated through adaptation of the measurement technique (longer integration times, rate-of-charge methods).

Conclusion

Unless further work proves successful, that should eliminate the present $\pm 1\%$ scatter in the scintillation detector counting rates, that detector seems to be inferior compared to the standard electron multiplier, on account of its lower reproducibility and its higher cost and complexity, for all applications involving ion currents in excess of 10^{-15} A.

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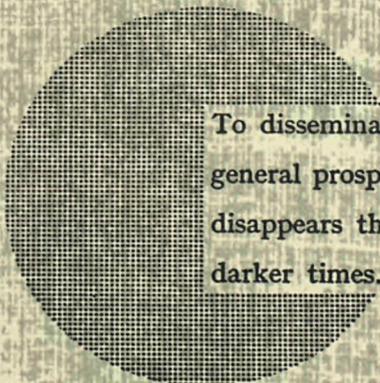
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