QUARTERLY REVIEWS

THE ELECTROMAGNETIC SEPARATION OF STABLE ISOTOPES

By R. H. V. M. Dawton, Ph.D., and Mervyn L. Smith, Ph.D., F.R.I.C.
(Atomic Energy Research Establishment, Harwell, Berks.)

Introduction

Early Developments in Electromagnetic Separation.—Ever since the demonstration by Sir J. J. Thomson, in 1912, of the existence of the isotopes of neon there has been continuous effort to obtain discrete samples of enriched isotopes for physical studies. A number of methods have been developed [1] but the electromagnetic separation method, which is the most versatile and has the largest separation factor, is the particular subject of the present Review. Aston [2] himself in 1922 considered this application of the mass-spectrographic principle, but Morand [3] in some early trials was unsuccessful and it was not until 1934 that successful instruments of this type were described. By this time there was great interest in nuclear studies so that isotopic targets were needed to give unambiguous results, and it was not surprising that almost simultaneous reports were made by Oliphant, Shire, and Crowther [4] who made ^{6}Li and ^{7}Li targets carrying up to 0·1 γ/cm.^{2} of element for proton- and neutron-bombardment studies, and by Smythe, Rumbough, and West.[5] These workers used a more efficient system with a magnetic lens and were able to obtain 1 mg. of ^{39}K (initial abundance 93·1%) substantially free from ^{40}K and ^{41}K; in 1937 they [6] obtained several mg. of the isotopes of the alkalis and were able to assign the natural radioactivities to ^{40}K and ^{87}Rb.

Yates [7] in 1938 used as the source a low-voltage arc into which the appropriate vapour was introduced, and this significant improvement enabled him to prepare nuclear targets of lithium, boron, and carbon. Walcher [8] developed a 90° machine with an ion beam of 100-cm. radius in a homogeneous magnetic field and collected about 0·1 mg. of the rubidium isotopes.

All these workers were using extremely small beam currents of the

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[1] C. J. Zilverschoon, Thesis, Amsterdam, 1954.
[2] F. W. Aston, "Isotopes" (E. Arnold & Co., London), 1941, p. 257.
[3] M. Morand, *Ann. Physique*, 1927, **7**, 164.
[4] M. L. Oliphant, E. S. Shire, and B. M. Crowther, *Proc. Roy. Soc.*, 1934, *A*, **146**, 922.
[5] W. R. Smythe, L. W. Rumbough, and S. S. West, *Phys. Rev.*, 1934, **45**, 724.
[6] W. R. Smythe and A. Hemmendinger, *ibid.*, 1937, **51**, 178, 1052.
[7] E. L. Yates, *Proc. Roy. Soc.*, 1938, *A*, **168**, 148.
[8] W. Walcher, *Z. Physik*, 1938, **108**, 376.

order of 100 μA and it was a most spectacular jump from their instruments to the large separators or "calutrons" developed in the U.S.A.[9] during 1942—1944 for the separation of ^{235}U from ^{238}U. In the autumn of 1945 it was possible in the U.S.A. to adapt these electromagnetic separation facilities for the production of enriched isotopes on the gram scale and since then the group at Oak Ridge have separated most of the 61 polyisotopic elements in the Periodic Table and built up stocks of 46 of them. Applications of these enriched samples in the U.S.A. have been numerous, as a recent 5-year summary [10] has indicated. Most of the consignments have been used for fundamental studies in nuclear physics, though the recent trend is for increasing application in the fields of general physics, chemistry, biology, and medicine. A brief account of the main types of application is given at the end of this Review.

Outside the U.S.A. a later start was made and gram quantities have only recently become available. At Harwell two machines were built,[11] the smaller prototype for the production of nuclear targets by direct deposition from the beam on to suitable backings, and the larger for the separation of gram quantities of enriched isotopes to provide a stock for research purposes. Recently, the smaller machine was closed down as it was of relatively low resolution and it was found better to make targets by indirect means using the stable isotope stock, but the larger machine has been put on a production basis, and so far 33 elements have been separated ; further elements are being separated as fast as possible. There are also in several countries smaller separators which are used for the separation of mg. quantities and for the preparation of nuclear targets by direct deposition. These instruments are described on p. 6.

So far, little has been published on the chemical aspects of this work and consequently a large part of the information given here is based on our experience with the Harwell separators. However, it has been possible to supplement this from the reports available of the U.S. work and also the reviews of Koch,[12] Bernas,[13] and Zilverschoon,[1] which, although dealing in most detail with physical problems, do also consider briefly the chemical ones.

Principles of the Electromagnetic Separation Method.—The electromagnetic separator essentially consists of an ion beam accelerated by suitable electric fields into a magnetic field where the masses separate by travelling in circular arcs of different radii and are then brought to a focus.

A practical form of the separator is a beam of ions passing through a sector-shaped magnetic field, the field being at right angles to the ion motion (see Fig. 1). The source of ions (A) is a line source perpendicular to the paper producing a flat beam which diverges in one direction with

[9] C. P. Keim, *Nucleonics*, 1951, **9**, 5.

[10] " Isotopes, 5-year summary of distribution ", August, 1951, U.S.A.E.C. [United States Atomic Energy Commission.]

[11] W. D. Allen, *Nature*, 1951, **168**, 451.

[12] J. Koch, Thesis, Copenhagen, 1942.

[13] R. H. Bernas, *J. Phys. Radium*, 1953, **14**, 34.

an angle of 2θ. This beam, after passing the magnetic field, is refocused in the region of B, the different masses being adjacent to each other.

The receiver unit consists essentially of a series of narrow boxes each shaped to receive a focused beam. In principle the design is straightforward but the cooling and the prevention of re-evaporation of the received materials frequently raise major difficulties, due primarily to the close spacing of the isotope foci (see p. 13).

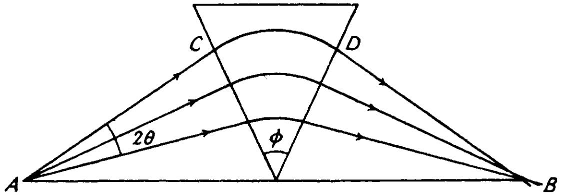

FIG. 1

Comparison with Other Isotope-separation Methods.—Recent reviews [1, 2, 12, 13] have considered and compared the several methods which have warranted large-scale development and these are:

(*a*) *Thermal diffusion.* This depends on the setting-up of a concentration gradient of molecules of different molecular weight when a temperature gradient is maintained in a gas or a solution.

(*b*) *Electrolysis.* The most important method for production of heavy water, and also may be important for other materials using ion migration in fused-salt baths.

(*c*) *Distillation.* Depends on the small difference in vapour pressure between molecular species of small mass differences.

(*d*) *Chemical exchange.* This potentially important method uses the fact that the equilibrium constants for a reaction carried out with different isotopes are slightly different. Consequently, if the reaction is carried out between two phases there will be a tendency for the isotopes to concentrate in separate phases.

(*e*) *Ultracentrifuge.* This method, which depends on separation in a centrifugal field, seemed to have attractive potentialities for high-speed separation, but the mechanical difficulties are considerable.

However, all these methods are alike in making use of very small separation factors, so that a large number of stages are necessary in order to obtain a highly enriched product. Thus, for any particular isotope a detailed process has to be worked out and then the appropriate plant designed and set up. In general, too, the methods are not well suited to the separation of intermediate isotopes of polyisotopic elements.

On the other hand, the electromagnetic separator is a very versatile instrument. In principle, it allows any isotope of any element to be segregated in one operation at very short notice since, as the separation factor is very large, considerable enrichment can be attained in a single stage. This universality is a unique feature and outweighs the limitations of rather small output and high primary cost of the installation so that the method is ideal for separating the small quantities of particular isotopes needed for nuclear research, and also is suited for obtaining gram stocks of isotopes for a variety of physical researches. It has even been found that the separator can compete with other methods for the production of certain high-activity tracer radio-isotopes (p. 21).

Factors affecting design of electromagnetic separators

Aberration of the Magnetic Lens.—This corresponds to the spherical abberration in optical lenses and can be corrected by shaping the boundary of the magnetic field, or by " shimming " the poles, *i.e.*, altering the gap where required. The lens strength, *i.e.*, its magnetic field, has to be held constant to about one part in 5000 parts.

Inhomogeneity of Particle Energy.—There are three main types of ion source : these utilise thermal ionisation, surface ionisation using the work-function effect, or electron impact : but the last, using 150-v filament electrons, is still generally favoured as it produces a high percentage of atomic ions from molecular compounds.

Thermal sources can be expected to have a very low energy spread; electron-impact sources have considerably more, say 30—300 v depending on their design. This spread broadens the foci (and, if excessive, would mix the isotopes) but the effect is less if high accelerating voltages are used : fluctuation of 50 v in the source is allowable for machines using 40-kv beams.

Space-charge Effect.—The mutual repulsion of the ions constituting the beam can also prevent the formation of sharp foci. It is noteworthy that, for strip beams, if the current per unit strip length is below a critical value even un-neutralised space-charge forces need not result in the broadening of the focus, although a displacement along the axis may occur. In practice, a considerable neutralisation of space-charge occurs due to electrons from the residual gas in the operating chamber so that focus impairment is slight.

Residual Gas Scattering.—Broadly, there are two types of collision encountered by an ion beam when passing through the residual gas of the vacuum tank. In one type the ion continues approximately in the same direction as a neutral particle (the charge having been transferred to the other body), and when such collisions occur over the range ACD (see Fig. 1) the particles are unlikely to enter the receiver. In the other case the ion retains its charge but becomes deflected (or scattered) from its original direction, and since most of this scattering occurs at angles of a fraction of a degree [14] this can cause an appreciable amount of isotope mixing at the receiver. This second type of scattering, because it reduces the isotope enrichment, is the more serious.

Machine Design Parameters.—The two essential requirements of a

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[14] N. F. Mott and H. S. V. Massey, " Theory of Atomic Collisions ", Oxford Univ. Press ; H. S. V. Massey and E. H. S. Burhop, " Electronic and Ionic Impact Phenomena ", Oxford Univ. Press.

separator are useful ion currents and sharp foci sufficiently spaced to take the receivers. The currents increase with the machine size and voltage. However, the sharpness of the foci is determined by gas scattering and no advantage is gained by increasing the ion-beam radius of the machine since the scattering rises proportionately. The most promising approaches are (i) reducing the amount of gas by improving the vacuum as far as possible consistently with stable operation, (ii) keeping path length small by making ϕ large (Fig. 1), and (iii) reducing the gas-scattering cross-section. Little is known on (iii) for small scattering angles but there is evidence from the operation of separators (see also Massey *et al.*[14]) that for a beam energy in the region of 10—50 kv the scattering is less at the higher voltages, and so an increase in voltage is beneficial.

A practical point is that it is difficult to fit individual beams of foci closer than 4 mm. in individual receiver boxes. (For masses in the region of 230 this calls for a beam radius of 90 cm.) Thus the receiver spacing determines the minimum radius (ρ) of the machine, while the output required determines the voltage and the length of the line source. The latter should preferably be less than $\rho/3$ for if it is too large the focus is impaired owing to the increased fringing of the magnetic field. Around these data the main vacuum tank and magnet can be designed. In passing, we may note that the cost of the magnet is almost independent of ϕ since, as ϕ increases, the maximum width of the beam is less.

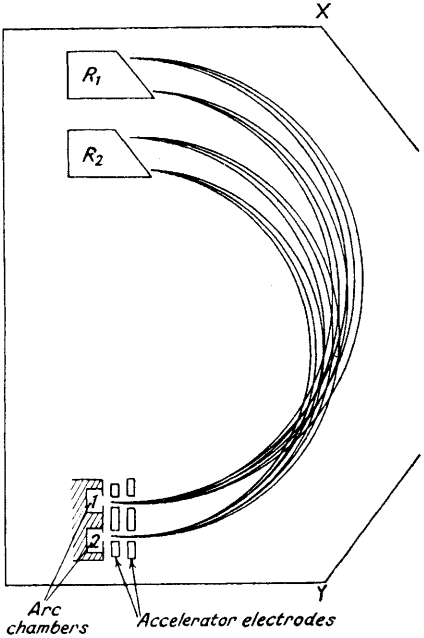

FIG. 2

Description of Harwell and other European separators

The Harwell Separator.—The machine used for most of the stable-isotope enrichment at Harwell was commissioned in January, 1950, two years after completion of the small prototype. In its design as much emphasis was placed on the through-put as on its enrichment factor. It is of the $\phi = 180°$ type (see Fig. 1) using magnetic shims parallel to the direction XY to achieve second-order focusing suitable for multiple-source operation (Fig. 2). This is somewhat similar to an early American machine at Berkeley, although there were numerous differences in design, *e.g.*, extensive use of graphite in the source units, adjustable spacing for receiver pockets, receivers designed for foil targets (*e.g.*, the covering of an area of foil with ^{10}B), and later, the use of tungsten heaters for source temperatures up to 1000°. The ion-beam radius is 2 feet with an enclosing vacuum chamber 7′ × 5′ × 13½″ evacuated by two 20″ oil-diffusion pumps. The two larger sides of this chamber are of 4″ thick iron to withstand the atmospheric pressure, and have bolted to them the magnetic "shims" for second-order focusing. The other four sides are of stainless steel.

Each ion source (see Fig. 3) consists in essence of a vapour chamber about 2 cm. × 2 cm. and 20 cm. long. When necessary, it can be heated to prevent condensation of the source vapour, the supply of which is discussed on p. 7. The ions are withdrawn through a slit 20 cm. long and of width 2—8 mm. according to the ability of the vacuum pumps to deal with the vapour, *e.g.*, for a non-condensable gas the slit would be narrowed. In the chamber the vapour has a pressure of 10^{-1}—10^{-2} mm. and is ionised by impact using 150-v filament electrons. Where higher source efficiencies are required an electron-reflection electrode may be used. Arc currents are usually about 2 A. Excitation by radio-frequencies has been tried on a small pilot plant, but unless the power is in the region of 1 kw the percentage of atomic ions from molecular compounds is too low.

Ions from the source are accelerated by two electrodes each having apertures about 20 cm. long and 2 cm. wide and designed to run hot (at a slight expense of the ion beam) to prevent the furnace vapour condensing upon them. The applied voltages are + 30 kv on the ion source, − 5 to − 25 kv on the first accelerator, and zero on the final electrode. (The weight of the whole source unit having twin arcs is about 700 lb.) The beam then traverses the main vacuum chamber where the pressure is in the region of 0·03 micron and after completing the 180° arc each isotope comes to a focus where it is received by a pocket. A total of 100 mA from each ion source has been obtained at these pockets, and careful design is needed in order to retain the isotopic material (p. 15).

A number of automatic controls are fitted. Contamination of the already collected material is prevented by a trip which operates when the collector currents deviate appreciably. Five seconds after such a trip the accelerating voltage is re-applied, and if the trouble was transient, *e.g.*, a spark followed by a power arc, the machine continues to run. If not, the process is repeated up to five times after which, if fault conditions still exist, the voltage is left off and an alarm bell rung. A time delay is fitted to the balance circuit to prevent just occasional sparking from tripping the high tension, although there is, in addition, a memory sensitive to too frequent sparking. The entire vacuum-system controls and the filling of the liquid-air trap are automatic. Stabilisation is applied to the magnetic field, the furnace temperature, and the arc current, the last by control of the arc-filament current. High-tension supplies, beside being highly stabilised, are also electronically power-limited.

Other European Machines.—In Europe at present the separators shown in the Table on p. 7 are in operation.

These machines are used mainly for the direct production of targets in association with accelerators rather than for gram-scale production of isotope samples for general scientific studies. The first two machines

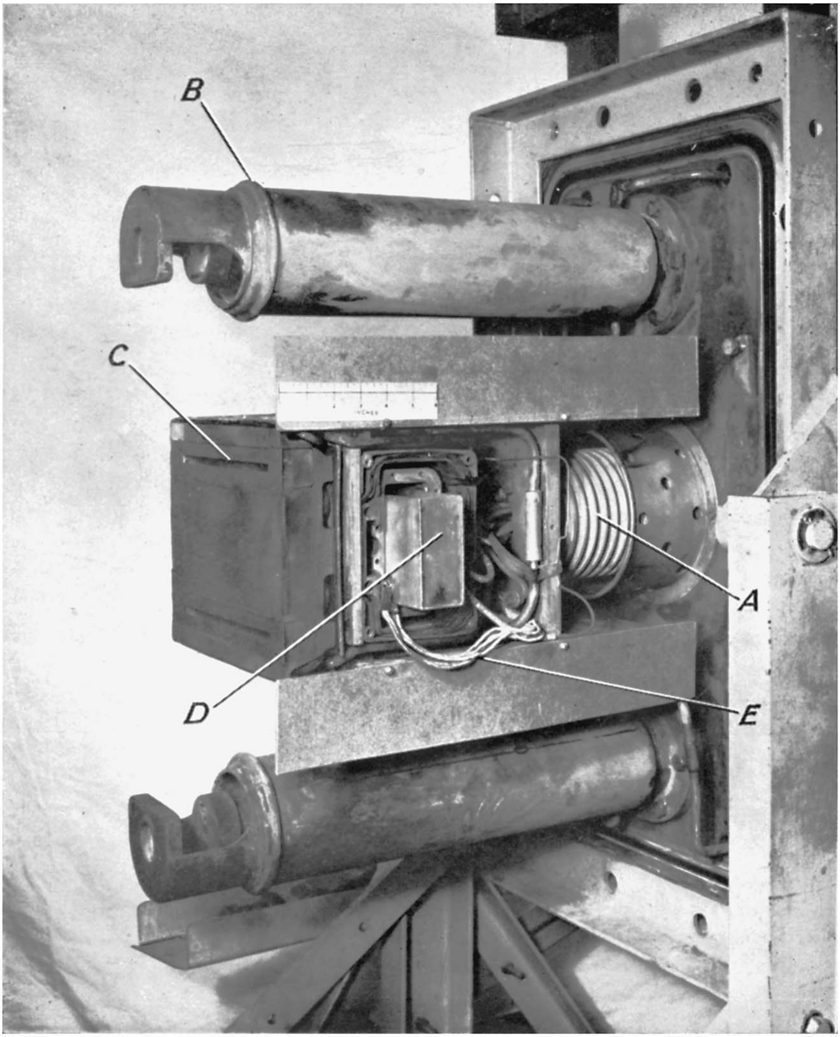

Fig. 3

Side-view of Harwell double-beam ion-source unit ; partly assembled ; electrodes and earthed shield omitted.

A, *Main insulator ;* B, *support for accelerating electrodes (upper beam) ;* C, *exit slit (upper beam) ;* D, *container for charge material ;* E, *thermocouple leads.*

Site	Ref.	Approx. date of first operation	Magnet type	Beam radius (cm.)	Max. focused beam current (mA)	Accelerating voltage (kV)
Copenhagen . .	15	1941	90°	80	1	70
Stockholm . . .	16	1948	90°	160	1	60
Amsterdam . .	18	1952	180°	100	10	25
Paris	13	1952	60°	50	10	35

give rather small beam currents but some valuable work has been done on preparation of targets of the rare gases [19, 30] (see p. 18). The larger machines in Amsterdam and Paris have been used to separate quantities of up to 100 mg. of various elements.

Chemical problems in the operation of the electromagnetic separator

Although the design and construction of a large separator are essentially a task for the physicist and the electrical engineer, yet the operation of the unit, dealing as it does with nearly all the elements in the Periodic Table, poses a considerable range of chemical problems, which may broadly be grouped under three main headings : (*a*) Selection and preparation of the source material. (*b*) Chemical refinement of the separated isotopes to the desired standard of chemical purity in a quantitative way without isotopic contamination. (*c*) Miscellaneous problems such as choice of materials of construction with the required electrical and mechanical properties under the conditions imposed by the temperature and by the corrosion effects in different parts of the equipment ; questions of toxicity and safety precautions.

Selection and Preparation of the Charge Material.—Criteria for selection. In order to obtain the ionised beam of the particular element being separated, it is necessary to feed into the arc chamber the element itself or a volatile compound at a vapour pressure in the region of 0·1 mm. Hg. Such source materials may conveniently be gases, volatile liquids having a vapour pressure of more than a few mm. at a room temperature, or suitable solids. The last are fed into the arc chamber from a furnace just behind it, and it is essential to arrange that there is an increasing temperature gradient from the source furnace up to the arc chamber to avoid any condensation.

The nature of the source material has an important bearing on the behaviour of the separator and in the preliminary selection three criteria are important.

(*a*) Solids must give a vapour pressure in the region of 0·1 mm. when held within the operating temperature of the source furnace, which for

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[15] J. Koch and B. Bendt-Nielsen, *Kgl. Danske Videnskab. Selskab.*, Matematisk-Fysiske Meddeledser, 1944, XXI, 1.

[16] I. Bergstrom, S. Thulin, N. Svartholm, and K. Siegbahn, *Arkiv Fys.*, 1949, **1**, 281.

practical purposes ranges from 100° to 1000°, though the Oak Ridge workers have recently developed a higher-temperature source unit.[17]

(*b*) The compound used must have a simple molecular structure with a minimum molecular weight. Quite commonly the solid element itself is available and when used has the considerable advantage that the non-ionised part of the emergent stream from the arc slit is condensed and so does not contribute to the pressure in the vacuum chamber and may, even, as in the case of calcium, act as a "getter" and so notably improve the vacuum. In default of the element, a halide is usually preferred since these compounds are simple and frequently have a convenient vapour pressure in the available temperature range. Oxy-salts are generally unsuitable as they give some oxygen-containing ions which can rapidly erode the tantalum or tungsten filament used in the arc source. Complex structures in general give side bands, though it is remarkable how much of the beam is composed of elemental ions even from quite complex halides such as WCl_6.

(*c*) The compound must be stable until it has all vaporised and should be as nearly as possible 100% volatile. An example of an unstable source material is silver iodide, which, although it is the most suitable from vapour-pressure characteristics, decomposes thermally, giving silver films which cause short-circuiting. It is, of course, necessary that there shall be no water of crystallisation present, and this commonly means that if the source material is very deliquescent it can be allowed only a minimum exposure to air. This requirement can make difficulties in operational handling, since the life of the filament in the arc source is commonly under 25 hours, so the source unit has frequently to be withdrawn from the vacuum chamber for refitting. Some materials, too, such as potassium iodide, decrepitate on heating, and this has the very undesirable effect of throwing small fragments of the source material forward into the region of the arc chamber and giving uncontrolled bursts of vapour and an unsteady arc. It has been found at Harwell that a considerable improvement in smooth running of the arc is achieved by melting the source charge in a vacuum before placing it in the separator, and this is now standard practice for difficult materials.

Review of source materials used. A useful list of source materials has recently been published by the Oak Ridge workers,[20] and various other workers have given some information. A recent exhaustive survey [21] of mass-spectrometer techniques is also of general value as a guide. As our own experience, too, is quite extensive this may be useful (see Table 1) for comparison with the published U.S. list.

The temperatures of the source, which are measured by a thermocouple clamped on to the outer surface of the heavy stainless-steel source container

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[17] W. A. Bell, L. O. Love, C. E. Normand, and W. K. Prater, *Bull. Amer. Phys. Soc.*, 1953, **28,** 5, 24; U.S.A.E.C., O.R.N.L. 1767.

[18] See C. J. Zilverschoon,[1] and J. Kistemaker and H. L. D. Dekker, *Physica,* 1950, **16,** 198.

[19] J. Koch, *Nature,* 1948, **161,** 566

[20] C. P. Keim, *J. Appl. Phys.*, 1953, **24,** 1255.

[21] M. G. Inghram and R. J. Hayden, U.S.A.E.C., A.E.C.U. 2120.

and are believed to represent the true charge temperature within a few degrees, correspond to vapour pressures of about 0·1 mm. The temperatures given by Zilverschoon and Bernas also agree quite closely with ours, though those quoted by the American workers are in some cases rather different.

TABLE 1. *List of source materials used at Harwell*

Source material	Source temp.	Total beam current * on collector (in mA), all isotopes	Source material	Source temp.	Total beam current * on collector (in mA), all isotopes
LiI . . .	650°	72	Zn . . .	450°	90
BCl_3 . . .	gas	25	$GeCl_4$ . .	gas	38†
O_2 . . .	gas	19	Se . . .	290	24
Mg . . .	560	57	RbI . .	610	25†
$SiCl_4$ . .	gas	48	Sr . . .	600	53
KI . . .	600	40	$ZrCl_4$ . .	140	28
Ca . . .	700	100	AgI . .	700	13
$TiCl_4$ . .	gas	13	$CdCl_2$ . .	510	26
$VOCl_3$ . .	gas	46	$InBr_2$ . .	240	32
$CrCl_3$ . .	650	90	$SnCl_2$ . .	270	40
$FeCl_2$ . .	600	60	Sb_2O_3 . .	490	32
$NiCl_2$ . .	610	60	$NdCl_3$ . .	910	20†
CuCl . .	390	55	WCl_6 . .	90	27†

* These are the currents obtained under normal conditions with a well-focused beam from a single-ion source.

† By use of a shortened ion-exit slit to achieve better resolution.

No problems exist in obtaining in pure form from ordinary commercial sources the several elemental samples which are listed above. In the case of the halides, however, it is necessary to prepare anhydrous samples from the commercial hydrates and this requires appropriate chemical treatment which can broadly be classified as follows :

(*a*) Stable anhydrous halides. These are melted in a vacuum before use, *e.g.*, potassium iodide.

(*b*) Anhydrous halides which can be readily prepared by drying commercial hydrated halides in an oven in a hydrogen chloride atmosphere to prevent hydrolysis, *e.g.*, $CuCl_2$, $FeCl_2$, $MnCl_2$.

(*c*) Liquid halides, *e.g.*, BCl_3, $SiCl_4$, $GeCl_4$, $TiCl_4$. Readily available, but it is important to specify a low free chlorine content to avoid difficulties with the gas in the arc. It may be necessary to redistil them before use.

(*d*) Halides which have to be specially prepared in the anhydrous state, *e.g.*, $ZrCl_4$, WCl_6, $MoCl_5$, most conveniently by direct reaction of the element with chlorine at an appropriate temperature. It is important that the metals should be oxygen-free before chlorination as otherwise some oxychloride will be formed.

(*e*) Rare-earth halides. These are difficult to dehydrate without some hydrolysis and the reaction has been studied by several workers.[22] Although the process is not difficult to carry out with small quantities, yet the equipment requires skilful design on the kg. scale to ensure a volatile product.

[22] D. H. Ahman, U.S.A.E.C., A.E.C.D. 3205, 1950.

In dehydration as much of the water as possible is driven off in an atmosphere of hydrogen chloride at a low temperature, after which the temperature is very slowly raised until the halide is anhydrous. This process is carried out at low pressure and gives completely volatile chlorides.

Apart from the halides there are some other special source materials such as carbon disulphide and various permanent gases.

There remain certain other elements, particularly those in the platinum and the palladium group, for which no convenient compound is available with a reasonable vapour pressure at medium temperatures. The U.S. workers have recently described [17] a high-temperature source unit in which the graphite charge container is heated by bombardment by the 35-kv electrons, and at a temperature of about 2500° these metals vaporise.

It has been pointed out by Keim [23] that the need for kilogram quantities of source materials of all polyisotopic elements has stimulated much chemical work, and he mentions particularly the separation of zirconium from hafnium and fundamental chemical studies on the rarer elements including the lanthanides.

Unfortunately, vapour-pressure data are not quite adequate for all the compounds used. Those available, apart from those in the standard reference books, are given for inorganic compounds in three recent compilations,[24] but there is some uncertainty in the published values for rare-earth halides and improved values have recently been published by Harrison [25] and by the Oak Ridge workers.[26]

Behaviour of Source Materials in the Arc.—Gas pressure in the arc and vacuum chamber. In the separator the operating conditions are markedly influenced by the character of the charge material, for a variety of physical or chemical reasons. For instance, it is necessary that sufficient gas should be in the arc chamber, otherwise the arc "goes into hash", *i.e.*, high-frequency oscillations are set up in it.[27] On the other hand, an excess of vapour will lead to breakdowns of the field and to scattering of the ions resulting in worsening of the focus.

A somewhat related difficulty is the influence of the gas pressure in the arc and in the main vacuum-chamber. When a complex halide is used, the number of volatile by-products of the chemical breakdown in the arc and the number of molecules which pass through without ionisation are appreciable. These will, at a constant pumping speed, increase the pressure in the main chamber and by causing scattering will again worsen the focus. This introduces a definite limit to the amount of source material

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[23] C. P. Keim, U.S.A.E.C., Y.662, 1950.

[24] D. R. Stull, *Ind. Eng. Chem.*, 1947, **39**, 540; L. L. Quill, "Chemistry and Metallurgy of Miscellaneous Materials", National Nuclear Energy Series, IV-19B, 196—207, McGraw-Hill, New York, 1950; R. L. Loftness, "Vapour-pressure Chart for Metals", U.S.A.E.C., N.A.A. SR. 132.

[25] E. R. Harrison, A.E.R.E. G/R 695, 1951.

[26] L. O. Love and W. A. Bell, U.S.A.E.C., A.E.C.D. 3502, 1950.

[27] D. Bohm, E. H. S. Burhop, H. S. V. Massey, and R. M. Williams, "Characteristics of Electrical Discharges in Magnetic Fields", National Nuclear Energy Series, I-5, McGraw-Hill, New York, 1949.

one can tolerate in the arc and hence in the final output of the separator (see Fig. 5).

A contrary effect is found with some elemental charges such as calcium, where, by reason of the condensable nature of the neutral material leaving the arc chamber and the " gettering " action, the pressure in the main chamber may fall to such an extent that the ion beam ceases to have sufficient space-charge compensation and defocuses.

Source efficiency.—There are a number of considerations which are important since they control the percentage of positive ions obtainable from an arc. First, there is the cross-section for ionisation of the particular element which depends also on the type of source used and on the running conditions, *i.e.*, the voltage drop from the filament to the arc chamber and from the current flowing in the arc. By increasing the electron emission from the filament, the efficiency can always be increased, but in practice this may not always be desirable as it may shorten the filament life too much. Apart from this, the number of " useful " ions produced depends on the complexity of the compound used ; *e.g.*, for WCl_6 the production of one W^+ ion may be accompanied by six Cl^+ ions which are lost as a side band. An average figure for efficiency of ionisation in the arc chamber has been suggested by Bernas [28] to be 10—20%, which means that over 80% of the molecules leaving the arc chamber are either neutral molecules or neutral dissociated atoms. Zilverschoon [1] has found for zinc that only 11% leave the source as charged ions (see Fig. 6). He has also reported on the mass spectrum of UCl_4 at normal operating conditions when the distribution is as follows :

Species . . .	U^+	UCl^+	$UCl_2{}^+$	$UCl_3{}^+$	$UCl_4{}^+$	Cl^+	$Cl_2{}^+$	U^{++}
Current, μA .	5700	900	500	250	50	2800	300	1100

Our experience has been similar with a variety of compounds but it is found that the extent of these side bands can be controlled to some degree by the arc adjustments available.

In the same paper are some interesting figures on the influence of gas pressure in the source on the emitted ion current and on the collector current. The results for argon are reproduced in Fig. 4, and it will be seen that, whereas the ion current reaches a maximum at a source pressure in the region of 3 microns, and then is fairly constant with increasing pressure, yet the collector current is very sensitive to the pressure in the vacuum chamber, for it attains a sharp maximum when the pressure is about 0·035 micron. Our experience with a number of elements has been very similar, a poor pressure in the vacuum chamber giving a low beam current at the collector and a poor focus owing to the gas-scattering effects. Some of our results are given in Fig. 5. These effects are very important, as Zilverschoon has commented, in controlling the collector current from different source materials. In general, gases and materials which give

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[28] R. H. Bernas, personal communication.

volatile end-products (such as chlorides and in particular the very volatile chlorides like BCl_3) whose unchanged molecules are gaseous, cause an increase in the pressure in the vacuum chamber and these materials give beam currents several times less than those achieved with solid elemental or condensable source materials.

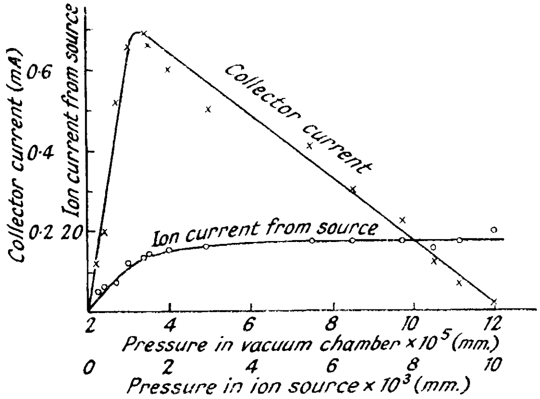

FIG. 4

Influence of pressure in source and tank on the emitted and collected ion currents with argon. [Reproduced, by permission, from Zilverschoon (ref. 1, p. 113).]

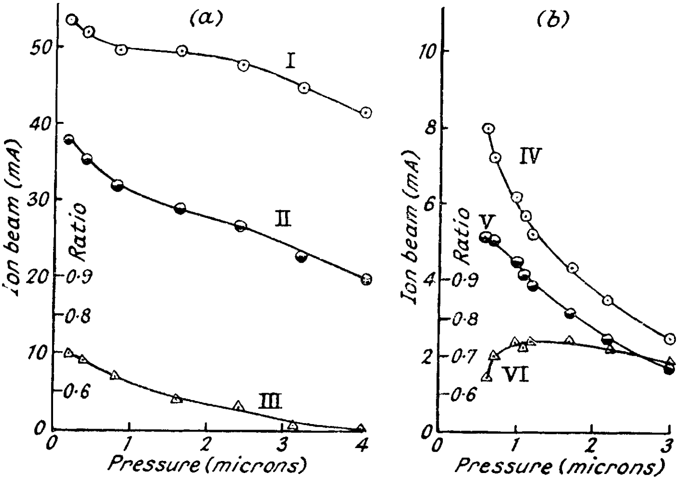

FIG. 5

Influence of pressure in vacuum chamber on collector-ion currents with various source materials and gases: (a) ^{115}In, *with* $InBr_2$ *as source;* (b) ^{40}A, *with gaseous argon as source.*

I, *Total beam,* $^{115}In^+$; II, *focused beam,* $^{115}In^+$; III, *ratio, focused beam : total beam.* IV, *Total beam,* $^{40}A^+$; V, *focused beam,* ^{40}A; VI, *ratio, focused beam : total beam.*

A difficulty which has been commented on by Bernas and by Zilverschoon, and which we have experienced, is connected with the deposition on the walls of the arc chamber of an insulating layer which prevents the discharge of the electrons and allows a negative potential to build up, causing instability and sparking in the arc chamber and a poor resultant focus.

This happens particularly in the case of reactive elements, such as Mg, Zn, Ca, with insulating oxides or nitrides, and occurs when the separator is opened to air in order to renew the filament. It is necessary to scrape the white deposit from the walls of the arc chamber in order to ensure smooth running; the effect can only be partly obviated by cooling the unit with an inert gas like argon before letting in air.

Another trouble due to the source material is deposition of the material, either in its original form or as an element from discharge of positive ions, on the accelerating electrodes in front of the arc source. This causes sparking and the resultant heating causes volatilisation of the material which results in more discharges until the whole machine finally becomes unstable. It is essential to keep the temperature of the electrodes well above the running temperature of the source and this is usually achieved with the help of a heater at the front part of the unit.

Experience has shown that the overall collection efficiency of the separator depends on control of all these factors but there are very few direct measurements on the true efficiency of the source and allocation of the causes of losses of ions at successive stages. However, Zilverschoon has given an analysis for the case of Zn and this is so interesting that it is reproduced in Fig. 6, from which it will be seen that the overall collection is only 1·8% of the weight of the charge used. The overall efficiency on the Harwell machine is rather higher than this and varies with different materials but we have no figures for the losses at various stages, and indeed these losses are likely to vary so much with controllable features of the unit that much experimentation would be needed to obtain useful results. This overall efficiency is not so important in terms of the cost of source material with most common compounds, but in the case of some rare elements and in the separation of artificially radioactive isotopes the percentage recovery achieved is, of course, a vital factor. The related question of retention in the pockets is considered below.

It is very difficult to get a real comparison between the behaviour of different elements in the arc because there are so many other variables connected with the machine and arising from the complexity of the source materials used. However, at Harwell a considerable range of experience is now available and Table 1 lists the optimum operational beam currents under stable running conditions for a number of elements. No analysis of these figures is attempted but the experimental values may be of general interest.

Problems at the Collector.—Collector design. At a single mass position in the Harwell separator the beam may amount to 100 mA and since this is accelerated at 30 kv the collector must dissipate over 3 kw at a line focus. It is not easy, in the limited space available, to provide sufficient cooling to ensure that the surface is kept cool enough to avoid

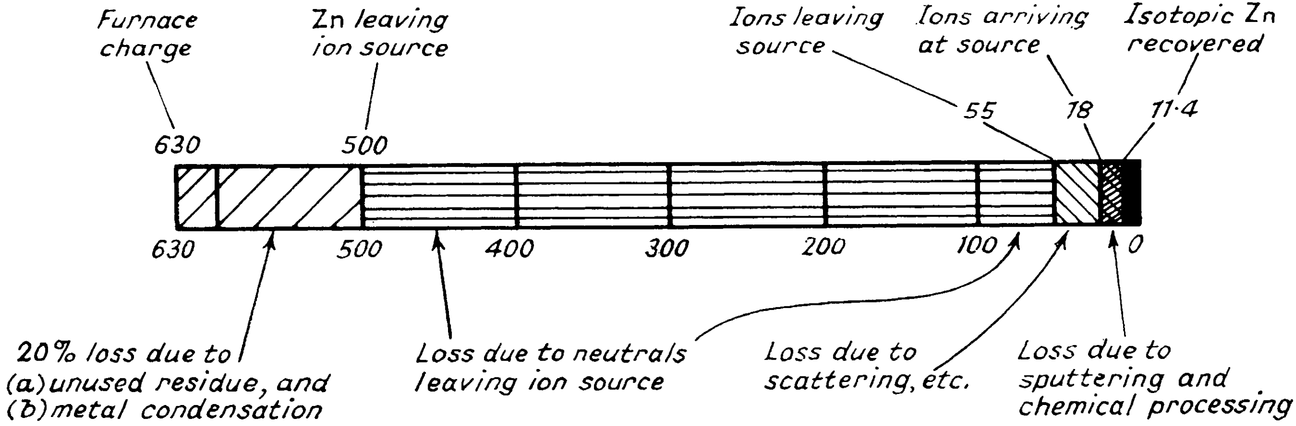

FIG. 6

Efficiency scheme for separation of zinc. [Modified from Zilverschoon (ref. 1, p. 123).] *All figures in mg./hr. Vaporisation efficiency* 90%. *Ion efficiency of source* 11%. *Overall efficiency of separator* 1·8%.

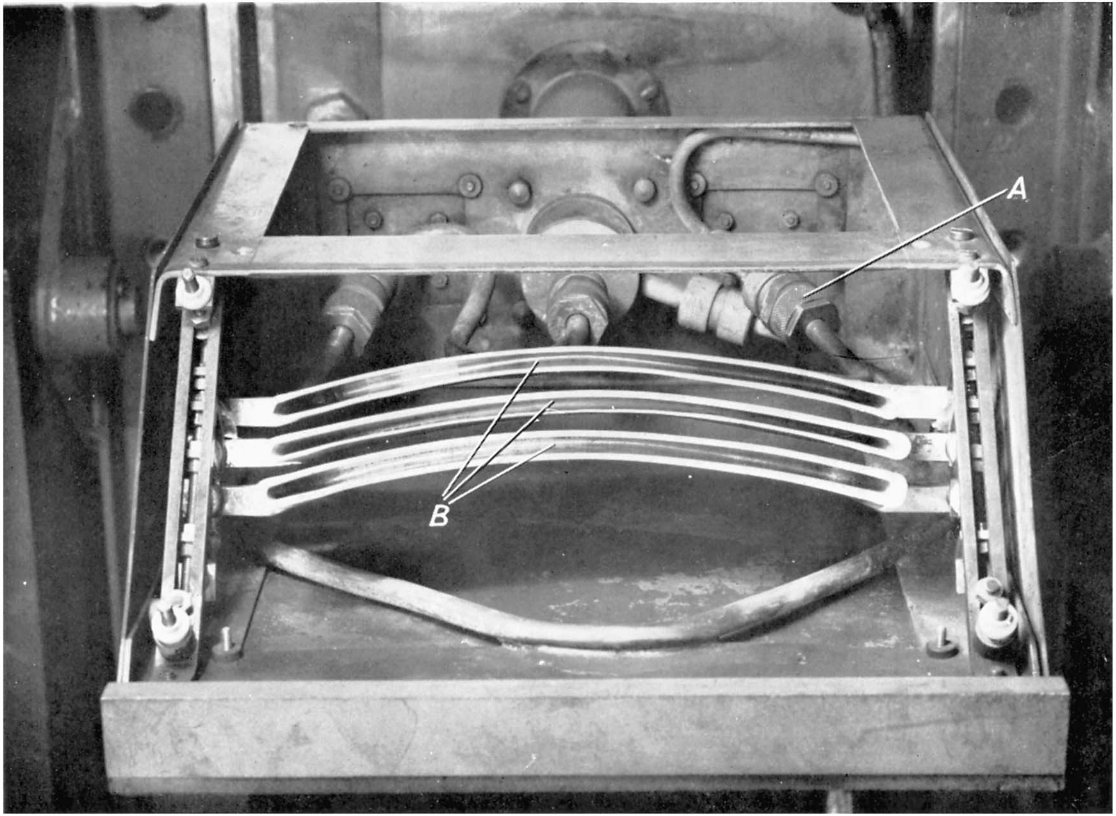

FIG. 7

Standard collector used at Harwell; front view.
A, *Water-cooling couplings;* B, *openings of individual collectors.*

subsequent re-evaporation of the collected isotope, and any failure of the cooling will mean, not only loss of the collection, but also melting of the collector. This problem is discussed in more detail in the next section. A further difficulty is that the ions of many elements bounce off the surface, either as ions or as atoms, and may be completely lost from a collector of unsuitable design. Moreover, material already deposited on the collector surface may be sputtered off by ions arriving later. These effects depend on the element being separated, on the element used for the collector, and also probably on the temperature which this surface reaches.

Most workers seem to have arrived at a collector design consisting essentially of a deep, narrow box, inclined at a slight angle to the beam so that the impact is on one face and reflected and sputtered material can be collected on the opposite one—Fig. 7 shows the detail of the Harwell arrangement. The material which bounces or is sputtered out hits the back surface of the front plate round the entry slits, where it stays providing it is not volatile at the rather high temperature reached by the uncooled front plate (not shown in Fig. 7).

In the Harwell separator it is found that up to 25% of the total collection may bounce back from the collector on to the back of the front plate. This material is, of course, enriched but, owing to cross-scattering, less highly so than material retained in the collector. Thus in a typical case with ^{6}Li the material in the collector was enhanced to over 99%, whereas sputtered material recovered from the front plate contained 89% of ^{6}Li. Zilverschoon, considering the same problem,[1] gives the possible loss at 30% from this cause, and Bernas [28] finds losses of the same order. Ion-deceleration electrodes have been tried at Harwell but the effect on sputtering seemed small; Bernas has also tried a system of this sort but it does not seem to be really justified. Another type of collector design is shown in Fig. 8. This has solid reception plates at the back of the pockets with a front surface at a sharp angle so that the reflected atoms are caught on the specially shaped walls of the collector. It is important to realise that the collector design is controlled by the total beam current expected, and in practice a compromise must be reached to give the maximum possible cooling and the maximum protection against cross-scattering in the space available. It is the aim to collect all isotopes simultaneously but this is not always possible on account of the small separation of foci, and if intermediate isotopes have to be rejected by allowing the beams to hit the front plate a further possibility of isotopic contamination arises since some of the local “ cloud ” of these atoms may be knocked into adjacent pockets by other atoms in the beam.

Another cause of loss is chemical attack by the atmosphere inside the vacuum chamber. For instance, if a chloride is being used as source material, the free chlorine may well attack the freshly deposited element if the temperature is sufficiently high and re-form the chloride which may then be volatilised off. Alternatively, there may be chemical exchange of the collected isotope with gaseous neutral compounds giving rise to isotopic contamination.

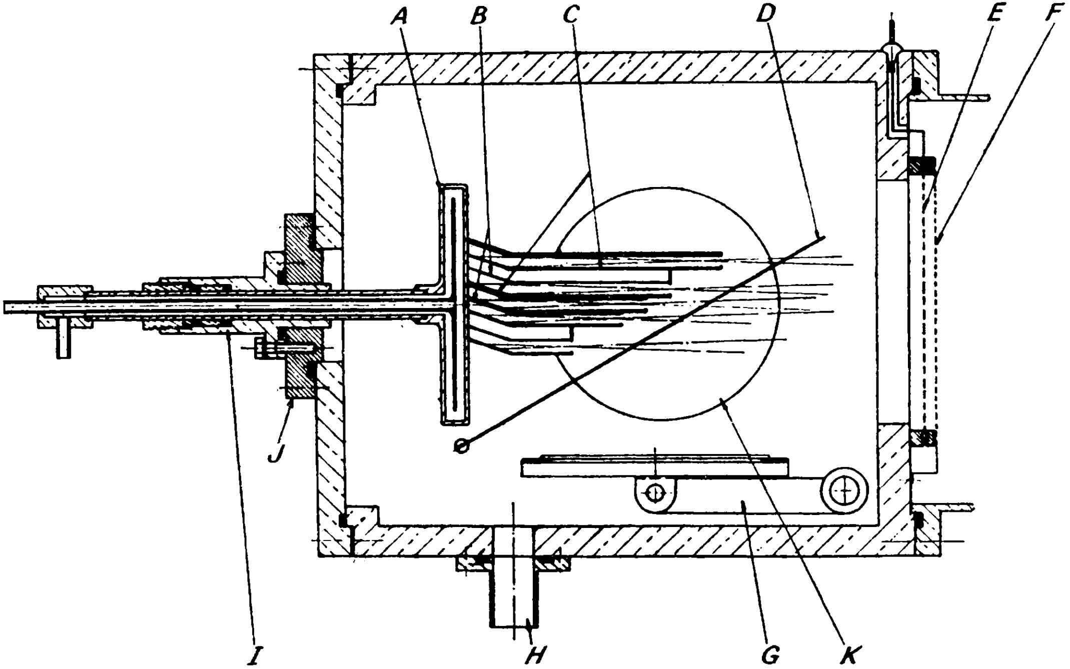

FIG. 8

Collector used by Bernas. [Reproduced, by permission, from ref. 13, p. 40.]

A, *Water-cooled support insulated by* J ; B, *receiving surfaces* ; C, *reflecting surfaces* ; D, *movable screen* ; E, F, *grids for space-charge neutralisation* ; G, *vacuum-isolating seal* ; H, *pump line* ; I, *Wilson coil* ; K, *Pyrex window at side.*

Collector cooling. The temperature which the collector will reach by the balance of power input, cooling due to radiation, and applied water-cooling is the most important consideration. It is readily calculated and provides the basis for design of the collector, the wall thickness, amount and position of cooling, etc., subject to the limitation of the space available and the fixed pocket separations. Where space permits it is best to solder cooling pipes on the outside of the top face of the collector where the beam hits ; alternatively, cooling is supplied at the back and reliance placed on conductivity of the collector material.

It is important that the surface opposite that on which the beam hits directly should be sufficiently cooled so that the vapour pressure of the deposited isotope will be appreciably below the pressure in the vacuum chamber, otherwise there will be a steady evaporation loss, and this requirement has been stated by Zilverschoon as follows :

" Vapour pressure of the material at the collector temperature for the rate of loss to be under 1% of the rate of supply should be less than

$$1{\cdot}8 \times 10^{-11} i\sqrt{(TM)}/A \text{ mm.}$$

where i is the current in the particular collector, T the absolute temperature of the collector surface, M the mass number of the isotope, and A the area of the front plate slit in cm.^{2}."

Sputtering.—Consider first a copper collector. In regard to their behaviour on hitting the surface the elements fall into two groups, those which build up on the collector surface where they first hit, such as Sn, Ti, Si, W, and B, and the great majority, such as Ca, Fe, Ni, Zn, Cd, which sputter the collector, cutting a deep groove, and are themselves thrown on to the opposite face intimately mixed with copper. Lithium seems to be an intermediate case. Elements in the first group form a surface scale more or less alloyed at the interface with the underlying copper ; this builds up in thickness until finally it peels off and falls away because of differential expansion. The collector has to be sloped back sufficiently to retain these separate detached fragments.

Elements which are sputtered are deposited, generally as an alloy, with copper from the collector surface on to the opposite, cooled, side of the collector. Thus a scale is built up containing only a few units % of the desired element until it separates from the underlying surface. Zinc behaves like this and the product has the characteristics of a low-zinc brass. Again, this process cannot be allowed to go on too long or the scale may bulge into the direct path of the beam, whereupon it overheats, melts, and loses some of its volatile constituent, an occurrence to which ^{40}Ca is especially prone. The sputtering effect increases with the temperature of the collector surface, so that with intense beams there is serious erosion and the collector life may be limited to a very few days. There is not much experience on this sort of sputtering, so the few figures that have been obtained on the Harwell separator may be of interest (see Tables 2 and 3). This order agrees approximately with that generally accepted as characteristic of cathode sputtering processes. On graphite the erosion is slower but the temperature may rise to red heat so this material is only suitable as a collector for the more refractory elements, such as W and Ti, which are then collected largely as carbides and oxides.

TABLE 2. *Ascending order of tendency to sputter of various target materials under positive-ion bombardment*

Al, Mo, W, stainless steel, Cr, Ag, Au, Cu

Retention of the more volatile elements. The elements are in a very reactive state when deposited, and, in fact, chemical effects have been used by several workers in the collection of elements such as sulphur and bromine, either of which can be held down on copper as compounds.

Mercury is a rather special case and has been separated at Oak Ridge [29] on water-cooled silver pockets. To prevent contamination of the separated mercury by random diffusion of non-ionised mercury the liner is also cooled to about − 50° with a special refrigeration system during the separation. Bernas has collected mercury on a copper collector cooled with liquid air.

TABLE 3. *Life of water-cooled copper collector with various bombarding ions*

Isotope	^{7}Li	^{40}Ca	^{56}Fe
Rate of wear, cm./1000 mA-hr. machine output .	0·008	0·21	0·18

A rather special problem is presented by the rare gases,[30] but the high-energy ions do penetrate into the first hundred atomic layers of a metal surface and, using this effect, Koch [19] has collected these gases at a concentration of several γ/cm.^{2} on silver, the amount obtained being probably roughly proportional to the accelerating voltage. Such tests have also been made at Harwell, and it has been shown that krypton is retained on aluminium at 100° but can be recovered by heating the metal *in vacuo* up to about 200°. Various metal surfaces have been tried and aluminium seems to hold the largest quantity of gas but, even so, the totals held are very small. Collection of larger quantities would necessitate either a rotating drum or a " roll-film " technique. Similar tests have been made on nitrogen, and here it seems that molybdenum has the best retention.

Chemical purity of the collector material. Both at Oak Ridge and at Harwell the collector materials generally used for intense beams are graphite and copper. Graphite, which can only be used for elements of very low vapour pressure, is very variable in chemical purity, being especially liable to contain Fe, Ca, Si, Ti, U, and Al in quantities up to a few hundred p.p.m.

Commercially available copper is very pure, but care is necessary in specifying any bonding materials—hard solders used in construction, or soft solders for fastening on the cooling pipes. At Harwell it is found

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[29] L. O. Love and W. E. Leysham, U.S.A.E.C., A.E.C.D. 3501.

[30] S. Thulin, *Arkiv Fys.*, 1955, **9**, 107.

very convenient to make the collector by electro-deposition on a stainless-steel former to the correct contour and wall dimensions. Such collectors are water-tight and present no foreign material to the ion beam.

Attention is also necessary to various constructional details of the collector assembly to make sure that no contaminating materials, likely to be troublesome, are in the way of either the direct or the scattered beams from which they may be sputtered into the pocket.

Chemical Problems involved in processing Isotopic Concentrates.—As will be apparent from the foregoing discussions the material as deposited cannot be used directly and an appropriate refinement process must be devised to give a stable, solid form of known mass and chemical purity. The procedures employed are essentially those of gravimetric analysis but a number of special considerations are involved, and it is probably more useful to elaborate these than to describe the details of particular refinement methods. Some of these problems have been discussed by the American workers.[31]

(*a*) *Impurities.* Apart from the expected isobars, a surprising variety of impurities are always found in the isotope collection. These arrive in various ways; vapour molecules in the vicinity of the collector may be knocked in by simple collision, or charged species formed at some point in the main beam by charge exchange may travel to the collector, together with doubly charged heavier atoms or complex species. Isobaric impurities which were of low concentration in the original source material may be highly significant in isotopes of low abundance. It is surprising how many atoms of impurities of quite different mass travel from the source area to the collector. Stainless-steel constituents are, for instance, usually found in any collection up to a concentration of several units %; potassium is a common contaminant of rubidium source material and arrives in a surprisingly high concentration in the collection of ^{87}Rb.

(*b*) *Scale of work.* The amount of the collected product is very variable, depending on the isotopic abundance, and may range from a fraction of a milligram to several grams. Thus procedures ranging from those of laboratory-scale preparations to semimicro-techniques are needed. Moreover, the work covers nearly two-thirds of the elements in the Periodic Table.

(*c*) *Removal of gross impurities.* In general, the amount of deposited isotope is quite small compared with the amount of collector material with which it is admixed. Thus zinc occurs as brass of 2—3% zinc content; graphite scrapings may contain only 1% by weight of the desired isotope. Hence the procedure adopted must be capable of recovering quantitatively a minor constituent of the crude collection. It is a nice consideration to decide how much of the collector surface to remove in order to be sure of recovering all the isotopic deposit without getting too great an excess of other impurities and risk of isotopic contamination.

(*d*) *Isotopic contamination.* To reduce this source of contamination the amount and chemical purity of reagents used must always be carefully considered, and appropriate vessels, especially in the ignition stages, chosen

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[31] A. J. Miller and B. S. Weaver, U.S.A.E.C., MDDC 1087.

for the handling. It has been found that the enrichment of the collection varies in different parts of the collector so that if sufficient mass-spectrometry service is available it is desirable to keep as separate batches scrapings from different parts of the collector. Further, in different runs materials of widely different quality may be obtained so that it is not wise to unite batches.

(*e*) *Solution of the collector product.* In some cases, *e.g.*, with boron and with metals such as titanium and tungsten, on graphite collectors, the material is not readily dissolved by the normal reagents. This difficulty arises because the deposit is put down in a high-temperature form or because combination with oxygen, nitrogen, or carbon at the collector surface gives very inert compounds.

(*f*) *Preparation of mass-analysis sample.* Mass analysis as well as chemical analysis is always required on the final product. Experience shows that at least 0·5 mg. is needed for processing to the chemical form required for mass analysis and this preparation is in itself a nice chemical exercise. It is usually necessary to ensure substantial absence of isobaric elements which might affect the accuracy of the mass analysis.

(*g*) *Form of the product.* A stable weighing form is needed, and for many applications a simple chemical compound is essential. For many nuclear targets, the element is most desirable so that it is best if at least a part of the collection of every isotope is available as the element. Hence, methods for making small quantities of the element without contamination are needed. It also frequently happens that the user requires a particular chemical form, so it is necessary to devise methods of making such conversions with minimum loss and isotopic contamination.

Materials of Construction and Corrosion Problems of the Separator.—These problems are highly specialised and can be divided into the following categories :

(*a*) *Source problems.* The source bottle must withstand the corrosive effect of the, frequently, molten source material, and, in the 180° machines, be non-magnetic. Further, the entire source unit also has to be at the necessary temperature to give the desired vapour pressure, while the high-vacuum system imposes further design problems. Fortunately, non-magnetic stainless steel is suitable for many source materials, including most molten halides and alkaline-earth metals. Graphite is extensively used at Harwell for various parts of the source structure, especially where hot halide vapours are passing, and is very satisfactory. Its chief limitation is found with vapours of the reactive elements. Thus, calcium forms carbides and causes swelling and distortion of the various fitted parts, though this effect is greatly reduced if these areas are kept sufficiently hot, say above 850°, so that calcium cannot condense.

Attack on the tantalum filament is mainly erosion by back-bombardment by positive ions, but it has also been found that if excessive pressures of some vapours such as WCl_6 or $ZrCl_4$ develop, these compounds crack thermally on to the filament and the resulting metal deposit alloys with, and soon weakens, the tantalum.

A further aspect of the work is concerned with selection of the ceramic parts, thermocouples, and the heat-shielding materials.

(*b*) *Construction of vacuum chambers.* Material for the main chamber must resist corrosion by the source-material vapours and by various scattered beams, have adequate mechanical strength, and be suitable for use in a high vacuum. At Harwell the practice is to use inside the main vacuum tank a stainless-steel liner to facilitate cleaning, and this has given very good service. Other systems have been described such as the tank at Amsterdam which has mild-steel pole pieces as side walls separated by brass plates pressed on to the rubber gaskets. The Paris separator has a composite construction of glass and copper tube.

(*c*) *Other corrosion problems.* The greatest general problem is concerned with the cooling-water supplies to magnet, source unit, liner, and collector. Here, copper and steel parts occur in the larger plants and can undergo serious corrosion. These risks are minimised at Harwell by the use of water treated in an ion-exchange unit to a conductivity of 15 micromhos per cm.^{3} and kept at a pH above 8.

Brief review of applications of electromagnetically enriched stable isotopes

Just as the discovery of isotopes led to an understanding of the average nature of atomic weights and paved the way to a comprehensive system of atomic structure, so the recent availability of stable isotopes in quantity for direct experimental work on specific nuclei is helping to establish a general theory of nuclear structure. Enriched stable isotopes are at present mainly used as targets in various types of accelerators in order to study specific reactions, but there are also many other types of study in which isotopes can greatly simplify the work and give positive identification to the nuclear species causing the effects observed. Recent short reviews by Allen [11] and by Keim [32] have described these studies, but it may be useful to list here some important fields of application to date. These include reaction studies ; neutron cross-section measurements ; natural radioactivity in, *e.g.*, ^{40}K, ^{87}Rb, ^{115}In, ^{147}Sm ; measurement of energy of fast neutrons (photographic plates with ^{6}Li) ; superconductivity ; optical studies of hyperfine structure and isotope shift ; preparation of high-activity radioactive tracers ; and stable tracers.

Space does not allow much consideration of such work but it may be mentioned that the neutron cross-sections are not only of theoretical interest to nuclear physicists but also are of considerable practical importance to the pile-power programme. Isotopic effects are normally only appreciable in nuclear interactions, but recent studies [11] with the tin and mercury isotopes have shown that the superconductive transition temperature T_c varies with the mean isotopic mass ; the theoretical prediction that $T_cM^{\frac{1}{2}}$ is constant has been approximately confirmed. In solid lattices one may not expect to be able to measure isotopic effects, but a recent X-ray diffraction investigation by Thewlis [33] has revealed differences of about 0·02%

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[32] C. P. Keim, *Ann. Rev. Nuclear Sci.*, 1952, **1**, 263.

[33] J. Thewlis, *Brit. J. Appl. Phys.*, 1952, **3**, 306.

between the lattice spacings of ^{6}LiF and ^{7}LiF. It may be that in some specially favourable cases isotopic effects may be shown in some other phenomena which depend in a rather sensitive manner on electron movements in lattices or on lattice distortion.

Radio-tracers of high specific activity, mainly for medical applications, are prepared by pile-irradiation of enriched isotopes. Thus ^{45}Ca is an important biological tracer, but when prepared by irradiation of natural calcium it has low activity since ^{44}Ca has an abundance of only 2%. Moreover, it contains undesirable activities of other calcium isotopes. By the use of ^{44}Ca enriched to about 98% a carrier-free tracer of high activity is obtained. In a similar way ^{59}Fe of high specific activity is prepared from enriched ^{58}Fe, whereas from natural iron the product from the 0·3%-abundant ^{58}Fe is of low activity and is also contaminated with the long-lived and undesirable ^{55}Fe. ^{51}Cr is similarly made from ^{50}Cr. Mention may also be made of the method of dating rocks by the use of ^{87}Rb. This natural radio-isotope decays to ^{87}Sr with a half-life of 10^{10} years ; by measuring the proportion of radiogenic ^{87}Sr to natural ^{87}Sr in rocks their age can be found. In practice, the analysis, involving extremely small quantities, is carried out by using the isotopic dilution technique, in which a definite amount of ^{84}Sr is added, and the ratio of this to the other strontium isotopes in the extract from the rock is determined in a mass spectrometer.

A recent survey [10] lists the stable isotope consignments which have been distributed from the U.S.A. and shows that the range of applications is rapidly widening. In this country so far the consignments have mainly been used for nuclear research, but now that an increasing range of stable isotopes is becoming available it seems likely that there will be a more general demand paralleling the trends in the U.S.A.

The Harwell work described herein was carried out by the stable isotope group, the engineering services, and the spectrochemical and the mass-spectrographic group.