ENERGY PER ION PAIR MEASUREMENTS IN PURE HELIUM

AND HELIUM MIXTURES

by

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ABSTRACT

A value for the average energy dissipated by an ionizing particle per ion pair produced for ^{241}Am alpha particles completely stopped in purified helium has been established as $43.5 \pm 0.5 \text{ eV}$ ^{on a relative basis.} Although this value differs from those previously reported by other researchers, indirect experimental evidence points to the conclusion that the purity of the gas was higher than that of other workers. Direct measurement of the water vapour impurity level has been carried out, together with indirect estimates of other impurity concentrations.

The accuracy of the absolute measurements of the energy per ion pair was primarily limited by the accuracy of the source emission rate measurements. These were carried out by 2π alpha particle spectrometry.

Sources emitting reduced energy alpha particles have been produced. The difficulties of calibration over 2π geometry have been investigated and have led to a study of energy straggling in absorbers. Ionization experiments using reduced energy sources at low gas pressure have proved unsatisfactory.

Gas mixture studies have been used to show that non-metastable excited states of helium must be responsible in part for the Penning ionization effect observed in contaminated helium, previously attributed to metastable states alone.

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1.1 Introduction

With the present increase of interest and importance being attached to solid state detection devices, it is often asked why fundamental ionization measurements in gases should be continued. The answer lies in the fact that a great deal of physics and chemistry concerning the processes involved in irradiated gases yet remains to be investigated. Many dosimetric techniques still rely on gas measurements, and the free-air ionization chamber, by which the Roentgen is defined, depends upon interpretation of results in a particularly complicated gas mixture. Other devices dependent upon ionization phenomena in gases range from pressure gauges to gas chromatography detectors. The elucidation of some of the remaining facets of the absorption of ionizing radiations in gases must be a worthwhile aim.

Ionization results from the bombardment of a gas by sufficiently energetic particles or photons. This is the phenomenon most easily detected and rightly considered to be of paramount importance in radiobiological philosophy. The point-heat theory of Dessauer (1923) and experimental determinations of ion density along the track of an ionizing particle were early attempts at providing a sound basis for accurate dosimetry on a microscopic scale primarily for radiobiology. Such measurements were usually made in the gaseous state. Little importance was then attached to the excited atom or molecule, although in some cases super-excited states of a molecule exist, in which absorption of energy in excess of that required to ionize the molecule does not result in ionization being produced (Platzman, 1962). It is the secondary processes involving excited atoms within the irradiated gas that will be of major importance within the work described in this thesis. In molecular gases, dissociation of the molecule or transitions of molecular excited states may also occur.

It is most pertinent to distinguish between primary and secondary events in energy absorption. The former occur as a ~~direct and~~ immediate result of the passage of the ionizing particle or photon through the gas, while secondary events arise as a result of the perturbed atomic or molecular states and will display a time differential from primary events. Excited atoms and molecules may have lifetimes of the order of 10^{-8} seconds

while metastable states are considerably longer lived. Such states are able to suffer many collisions during their lifetime, a situation which has produced surprisingly erroneous results in ionization measurements in helium. This arises because of the relatively high energy of the helium metastable states. A collision of an atom in a metastable excited state with any impurity with an ionization potential less than the energy of the excited state will result in the impurity being ionized. Another long lived component in an irradiated gas is the subexcitation electron which may suffer many collisions before becoming attached to a gas molecule.

Interactions within an irradiated gas are exceedingly numerous and complex. Experiments may be designed specifically to investigate some of the individual fundamental events but in considering the overall effect of ionizing radiations it has been usual to measure the efficiency of ionization by determining the average energy required to form an ion pair in the gas. This is given the symbol 'W' with units of eV per ion pair. Literature referring to W values is now considerably voluminous. The value of W for all gases lies between 25 and 50 eV. From the detection sensitivity point of view, this compares unfavourably with solid state detectors in which W is of an order of magnitude less. Dependence upon particle type and energy in a particular gas has been observed, but the difficulties in assessing the results from a particular experiment are often considerable. The design of experiments to investigate phenomena due to low energy primary particles is not easy.

1.2 W values in gases

W is defined by I.C.R.U. (1962) as equal to E/N_w where N_w is the average number of ion pairs formed when a charged particle of initial energy E is completely stopped by the gas. Ions arising from the absorption of bremsstrahlung emitted by the charged particles are not counted in N_w . Since W depends upon the energy of the charged particle, a differential W may be defined as $W_{\text{diff}} = \Delta E / \Delta N_w$ where ΔE is a small energy loss by the primary particle along its track, and ΔN_w is the corresponding increment of ionization accompanying this energy loss.

An accurate knowledge of W or the variation of W with particle type and energy is of critical importance in many aspects of radiation dosimetry which involve the conversion of the amount of ionization occurring in a gas to the amount of energy absorbed by the gas, and, further, by the Bragg-Gray relationship to the energy absorbed in solids. A special case is the conversion factor of rads per Roentgen for air exposed to X and gamma rays.

Three parameters are significant in W value measurements, the type of gas, the type of particle used to ionize the gas, and the energy of the primary ionizing particle.

With regard to the gases used, it has been shown that to a first order of approximation W is independent of the type of gas and is about twice the mean ionization potential. As may be seen in table 1.1, the ratio of W to the mean ionization potential is about 1.73 for the noble gases (these being the only monatomic gases which have so far been studied). For polyatomic gases, this ratio is higher because an ionizing particle may lose energy by more processes in these gases than in monatomic gases.

TABLE 1.1 SOME W VALUES AND IONIZATION POTENTIALS (Fulbright, 1958)

GAS	MEAN ION. PTL. (I)	$W_{\text{electrons}}$	W/I	W_{alphas}	W/I
HYDROGEN	15.4	36.9	2.40	36.6	2.38
HELIUM	24.6	41.3	1.68	44.4	1.80
NITROGEN	15.5	34.9	2.25	36.3	2.34
OXYGEN	12.2	31.3	2.57	32.1	2.63
NEON	21.6	35.9	1.66	36.8	1.70
ARGON	15.8	26.3	1.66	26.3	1.66
KRYPTON	14.0	24.4	1.74	24.1	1.72
XENON	12.1	22.1	1.83	21.9	1.80
AIR	-	34.2	-	35.2	-
CARBON DIOXIDE	13.7	32.7	2.39	34.1	2.49
METHANE	13.1	28.1	2.15	29.1	2.22
ETHYLENE	10.6	26.4	2.49	-	-

There remains a great deal of work to be carried out, particularly in the field of gas mixtures. One such mixture is air, in which W was thought to have been established to within 0.5% until recent studies at the National Bureau of Standards (Chappell, 1967) showed possible discrepancies. These were attributed to possible variations in the composition of air. Recent work under the direction of the author on this topic at the University of Surrey (Evans, 1970) has tended to disprove this hypothesis. Tissue equivalent gas is also being studied in the same research programme. The facility for determining the composition of the mixtures is being provided by an analytical mass spectrometer. The special problem of impurity levels in mixtures will be considered in a later chapter.

Most accurate experiments have been carried out using particles emitted by radioactive isotopes. These have certain inherent limitations. Alpha particles with a definite range in gases provide satisfactory sources, particularly since their line spectra is usually uncomplicated by unwanted beta radiation although gamma radiation is often present. One obvious limitation is the available energy range between about 4 and 9 MeV, although attempts to produce reduced energy sources by the use of absorbers have been made (see section 3.4.4.3) and permit some information to be gained regarding energy dependence. A more fundamental objection is that in gamma ray dosimetry the ionization is produced by electrons rather than by alpha particles, and in neutron dosimetry protons and other recoil particles are the important primary particles. The use of W values for alpha particles in X, gamma, and neutron dosimetry is not, therefore, justified without prior validation. Careful experiments by Jesse's group showed that the ratio of W for alpha particles to W for beta particles differs from unity by only one or two percent in the case of the noble gases and hydrogen, but by about 6% for molecular gases. Since W for alpha particles tends to W for beta particles as the energy of the alpha particle increases, it is reasonable to suppose that the reason for this discrepancy lies in processes occurring near the end of the alpha particle track.

The determination of W for electrons and beta particles is more difficult than for alpha particles because of their greater range, and because of the complex energy spectrum of the beta particles emitted by radioactive isotopes. Most groups have made use of low energy beta sources

such as ^3H , ^{14}C , and ^{35}S . The use of low energy electron guns having the advantage of monoenergetic particle selection has not yet been exploited, although Barber (1955) has used a linear accelerator as a source of high energy electrons.

Recoil protons with implications for neutron dosimetry have not been thoroughly studied although some results in the 2 MeV region indicate that W is very similar to that for alpha particles. Heavy recoil nuclei have been the subject of some limited studies (Jesse, 1956; Stone, 1957) but the current increase of interest in heavy ion accelerators should permit an extension of the data. Phipps (1964) has shown that, for a given velocity, the value of W increases with the mass of the ion. Some fission fragment studies have been carried out by Schmitt (1956). With the accelerator facilities now becoming available at the University of Surrey, preparations are well advanced for a programme of research initially limited to a study of low energy protons and alpha particles.

The classical work on energy dependence of W has been carried out by Jesse (1950, 1955a, 1961) using alpha particles. His results indicated that W is independent of energy for argon over the energy range of about 1 to 9 MeV, whilst for polyatomic gases it decreases with an increasing energy approaching the value for W_{beta} at high energies. More recently, Chappell and Sparrow (1967) have carried out experiments which suggest that W is constant in argon for alpha particles over the range 1.08 to 4.188 MeV, but that nitrogen and air show an increase of about 1% over the same energy range.

The assumption of a constant W for alpha particles in argon has been challenged by Leake (1967) who was concerned with the measurement of W for recoil nuclei in carbon dioxide. By regarding W as a function of particle velocity rather than energy, he was able to conclude that the W values of Jesse for 1.02 MeV alpha particles require an increase of 1%.

W for beta particles is evidently constant down to very low energies. Although experimental data is difficult to produce, Jesse and his co-workers (Jesse, 1958) have carried out measurements with beta particles of tritium and other isotopes and have found no variation in W_{beta} . Rajewsky and

Lang (1961) extended this work to lower energies using X-rays down to 6 keV. An increase in W was observed at very low energies. A useful review of this and similar work has been carried out by Whyte (1963).

1.3 Historical survey of W measurements in helium

W measurements in helium have been carried out for a number of years and wide discrepancies unaccounted for by experimental inaccuracies have been noted. Table 1.2 shows some of the results produced. It is interesting to note that little work has been carried out on the energy dependence of W in helium.

TABLE 1.2 SOME W MEASUREMENTS IN HELIUM

AUTHOR(S)	IONIZING SOURCE	TYPE OF ION COLLECTION	W (eV per ion pair)
Valentine and Curran (1952)	^{210}Po	slow	31.7 ± 0.6
Jesse and Sadauskis (1953)	^{210}Po	slow	42.7 ± 0.5
Herwig and Miller (1954)	natural uranium	fast	44.2 ± 0.9
Bortner and Hurst (1954)	^{239}Pu	slow	46.0 ± 0.5
Haerberli et al. (1953)	^{210}Po	slow	29.6 ± 0.3

The assumption that total ion collection is achieved has long been a matter of concern. Although the use of extrapolation techniques to determine the true saturation ionization current have been questioned, they are still generally accepted as being a useful guide to collection efficiency. Noble gases do not suffer from the disadvantage of undergoing appreciable electron attachment.

While the impurity effect in helium (Penning ionization) was first observed by Jesse in 1952 (Jesse, 1952), there was no work aimed at determining the actual purity of the gas used. Chemical and physical methods of purification were used and the expected decrease in ionization current observed. When the ionization had reached a minimum, it was assumed that the gas was pure. Since it has been shown by several workers that continuous

repurification is necessary, it is evident that a state of dynamic equilibrium exists between the removal of impurities by the purification system and the diffusion into the gas of unwanted impurities either by outgassing of the apparatus or by leaks. There is little in the literature to suggest that this effect has been seriously considered, and it was the initial purpose of the work described in this thesis to investigate the purity of the gas. Jesse considered the problem by adding small quantities of impurities to the helium which he assumed to be pure. However, since his W values for 'pure' helium are lower than those of some other workers, it appears that his original gas was not wholly pure. This must make his impurity mixture experiments of limited quantitative value.

Only one measurement for helium has been recorded using the fast method of electron collection (Herwig and Miller, 1954). These workers recorded a W value somewhat higher than those produced by slow collection methods, and suggested that this could be a truly physical effect involving differences in collection times. Later work (Bay and Pearlstein, 1963) has tended to disprove this hypothesis. It is true that the influence of a Frisch grid is liable to introduce errors and could lower the efficiency of electron collection, although some attention must also be paid to the conflicting possibility of electron multiplication in the region of high field strength near the wires. As yet, no fully satisfactory explanation of the different results has been produced.

1.4 Helium gas mixtures

Studies relating to ionization production in gas mixtures can be divided into two distinct categories. The first of these, in which the constituents of the mixture are a considerable fraction of the total, have been investigated usually as binary mixtures in order to examine the ultimate distribution of energy between each gas (Moe, 1957; Valentine, 1958). The second category of mixtures is that in which a trace of additive gas exerts a profound influence upon the behaviour of the mixture. This effect, called the "Jesse effect" following the original investigations by Jesse (1952) is extraordinarily pronounced for helium. Jesse showed that additive concentrations of less than 0.1% produced an additional observed ionization of about 50%, the actual figure being dependent on sub-excitation electrons which have a long lifetime (Platzman, 1955).

Ionization by alpha particles in helium may be produced by the following two reactions:

The inelastic scattering process represented by (1) produces electrons, the majority of which are sufficiently energetic to produce secondary ionization as in (2). Excited helium atoms are produced by the following reactions:

It has been shown that approximately half the total excitation is in the 2^1P state (Bennett, 1962). Platzman (1955) has calculated that about 90% of the metastable excited states produced are in the singlet state since direct production of the triplet state by alpha particles is forbidden. Some triplet states may be formed by energetic secondary electrons.

As a consequence of the high pressure and large dimensions involved, very little vacuum ultraviolet resonance radiation will escape to the walls of the chamber (Holstein, 1947). In the absence of collision effects, excited $n^1\text{P}$ states would lose energy through radiation and fall to the 2^1S metastable level. The lifetime for such a reaction is about 10^{-7} seconds (Bennett, 1962). At 1000 mm Hg pressure, these excited states make about 10^3 gas-kinetic collisions with ground state helium atoms during this time. Reactions of the type:

where $n = 3, 4, 5$ and He^* is another excited state with the same principal quantum number as the initial ^1P level, occur with cross-sections between 10^{-15} cm^2 to 10^{-13} cm^2 (Maurer, 1940). Two body collisions leading to molecular ions as shown in the following reaction are important in the higher excited states of all noble gases:

There is little doubt that Penning ionization is responsible for the increase in ionization observed by Jesse in his study of small concentrations of impurity in pure helium. The results he obtained are shown in figure 1.1. The Penning reaction, as shown in (7) is energetically possible provided that the helium excitation level exceeds that of the ionization potential of the impurity, X.

Figure 1.1 The Jesse effect in helium

It has been usual to attribute the effect to metastable excited helium atoms which have a lifetime of about 10^{-3} seconds. The 2^1S and 2^3S singlet and triplet states have energies of 20.6 and 19.8 eV respectively. These, and some other important energy levels of helium are shown in figure 1.2.

Figure 1.2 Some important energy levels in helium

Experiment has shown (Jesse, 1955b) that neon, with an ionization potential of 21.6 eV, does not exhibit an impurity effect in helium whereas other common impurities with lower ionization potentials do. This is consistent with the hypothesis that metastable excited states are responsible for reaction (7). If molecular impurities are examined, molecular dissociation will usually predominate over ionization.

Some non-metastable n^1P states may also produce Penning ionization if during their lifetime they encounter an impurity atom. The probability that a two body collision will occur is pressure dependent but, since at 1000 torr an n^1P state makes about 10^3 collisions with ground state atoms in 10^{-7} seconds, it would be expected that impurity concentrations of the order of 1 part per 1000 would result in Penning ionization by n^1P states. The available transitions for an n^1P state may be summarised as follows:

1.5 Theoretical calculations of W in helium

Helium is a gas which has been favoured for calculations of ionization yield because of its relatively simple structure. W values for gases were first calculated by Bethe (1930) and improved by Bagge (1932), but Fano (1946) produced the first value for helium, correctly predicting a much higher value than the then currently accepted 30 eV per ion pair. It is now known that impurities were responsible for the poor observations at that time.

A collection of values calculated by various workers is shown in table 1.3.

TABLE 1.3 THEORETICAL CALCULATIONS OF W IN HELIUM

AUTHOR(S)	W (eV per ion pair)
Fano (1946)	38
Erskine (1954)	41.1
Platzman (1955)	42.2 ± 2
Miller (1956)	46.5
Peterson and Green (1968)	46.0
Alkhazov and Vorob'ev (1969)	46.6

The discrepancies between calculated values arise partly because of the assumptions necessary to perform the calculations and partly because of the use of poor data. Erskine improved upon Fano's approximate derivation by considering a number of alpha particle energies and the change of excitation and ionization cross-sections with energy. Among the assumptions allowing a calculation of the mean value of W are the independence of W on particle type, and, to a lesser extent on energy. The lower energies are treated thoroughly in the recent work but there is a lack of experimental data in this region for light ions and electrons. Most authors have neglected some of the relatively unimportant processes including elastic collision losses, production of doubly charged atoms, and capture and loss of electrons by alpha particles. While these processes may well be insignificant, particularly at an alpha particle energy of several MeV, the formation of singly charged helium molecules, He_2^+ , is quite important at pressures above a few tens of torr (Platzman, 1955). Since theoretical calculations of W have invariably neglected this phenomenon, the recent values of about 46 eV per ion pair are probably too high by about 3 eV (Platzman, 1961).

The resulting good agreement with the observations to be described in this thesis may well prove fortuitous; additional confirmation should be sought, particularly regarding the influence of molecular ions.

2.1 Introduction

The purpose of this short discussion is to define the reasons behind and subsequent development of the experimental programme discussed in this thesis.

The significance of helium molecular ion formation has already been discussed in chapter 1. Platzman (1962) suggests that, since the reaction is pressure dependent, the apparent energy per ion pair for pure helium will increase some 6% as the pressure is reduced from atmospheric pressure to about 10 torr. The appearance potential of He_2^+ has been established by mass spectrometric techniques (Hornbeck and Molnar, 1951) and optical spectra also demonstrate the presence of the ion (Bennett, 1962). Theoretical calculations of the energy per ion pair in helium, which ignore molecular ion formation, also tend to support Platzman's hypothesis when compared with experimental results at relatively high pressures but a direct observation of the effect in a total ionization experiment has not been carried out. Accordingly, an experimental programme was set up with the aim of elucidating the pressure effect in helium.

2.2 Experimental system

Historically, the work of Bortner and Hurst (1954) has been assumed by some workers to be the best available data for the energy per ion pair in helium, an unfortunate consequence perhaps of the tendency to select the highest observed value in view of the significant sensitivity to impurity concentration. Also, the experimental arrangement of Bortner and Hurst made little demand on special source preparation, and so a similar parallel plate ionization was designed for the present work, in which alpha particles from a planar source would be completely stopped within the gas volume. As an improvement on previous designs, the ionization chamber and gas purification system were both arranged to allow baking at 200°C under vacuum for extended periods in order to reduce outgassing of impurities.

Resulting ionization was to be collected and measured using an electrometer. An absolute value for the energy per ion pair could be calculated from a knowledge of the ionization current and the total energy absorbed by the gas in terms of numbers and energies of particles emitted from the source. A schematic diagram of the ionization chamber system is shown in figure 2.1.

Figure 2.1 Schematic diagram of the ionization chamber system

An electrode separation of 15 cm was chosen as the maximum practicable using a 2π source. The Bragg-Kleeman Rule indicates that for complete stopping of alpha particles within this range at a pressure of 10 torr of helium, it would be necessary to reduce the energy of natural alpha particles to a few tens of keV. The problems of suitable source production and measurement are discussed in the following chapters. It is evident that in this region the energy dependence of W in helium is of particular significance.

2.3 Experimental programme

The initial aim of the experimental programme was to re-examine the

energy per ion pair value in pure helium at about atmospheric pressure, bearing in mind the currently accepted values. Various types of alpha particle source were considered with the design limitation that a planar source should be used. The difficulties of calibration of source emission and energy emission rates are described in chapter 3. Two publications have already resulted from this work (Yeates, Harris, Doust, and Parish, 1969; Yeates, Harris, and Doust, 1970) and are included as appendices 1 and 2 to this thesis. They suggest the possibility that previous workers have not thoroughly considered the problems arising.

Following the construction and commissioning of the ionization chamber and associated vacuum equipment, a gas purification plant was also designed and constructed. The leak tight integrity and outgassing rate of the complete system was tested in a series of experiments.

A conventional ionization current measuring system was used, based on a vibrating reed electrometer. Particular attention was paid to devising a novel automatic arrangement which allowed unattended operation of the equipment.

An energy per ion pair value for helium obtained by reference to standard gases with well established values indicated that the apparent value did not agree with currently accepted figures. Since most systematic errors were eliminated by the comparative measurement, the significant remaining inconsistency between various workers was that of gas treatment and purity. It will be argued later that the greater gas purity achieved meant that the results obtained in this thesis are more accurate than those previously reported, and much experimental evidence from both direct and indirect gas purity measurements will be presented to support this claim.

Evidence from a study of gas mixtures was obtained which indicates that metastable excited states alone are probably not responsible for the impurity effect in helium. This confirms the theoretical predictions of Erskine (1954).

The intended investigation into pressure dependence proved to be impracticable due to the energy limitation on ions entering the gas.

It may be possible to achieve a reduced energy source to the specification required but this is not straightforward, and in any case calibration of such a source would be extremely difficult, a consequence of the uncertainty of the energy dependence of the energy per ion pair in any gas. It would almost certainly be better to employ either monoenergetic electrons from an electron gun or ions from a positive ion accelerator.

3.1 Introduction

In any ionization experiment, careful attention must be paid to the ionizing source. With the value of W dependent upon particle type and energy, it is desirable to use a monoenergetic source of charged particles such as may be obtained from an accelerator. The technology of accelerator beam production and measurement is not to be considered here but a brief discussion of work that has been carried out in preparation for experiments using the 2 MeV Van de Graaff accelerator at the University of Surrey is given in section 3.6. Much useful work remains to be done with, for example, protons of various energies obtainable from an accelerator. Such studies could have ~~far reaching~~ consequences in the field of neutron dosimetry.

Many workers have used radioactive isotopes, chosen to suit their individual requirements both in terms of the type of radiation emitted and its energy. Some of these isotopes, their properties, and the types of mounting used, will be discussed in the following two sections. Since the present work is concerned only with alpha particles, discussion will be limited to alpha emitting isotopes.

3.2 Alpha emitting isotopes used for W value measurements

It is advantageous to use an isotope which emits only alpha particles and this is most closely approached by ^{210}Po . However, the comparatively short half life of this isotope makes decay corrections necessary. An additional difficulty lies in the reputation of polonium to "creep" (Bagnall, 1962). The geometric conditions of the present work closely approach those of Bortner and Hurst who used a planar source of ^{239}Pu . Both ^{239}Pu and ^{241}Am emit some low energy gammas but internal conversion is greater in the case of ^{241}Am . The choice of natural uranium by Herwig and Miller could only be justified in their use of a pulse chamber in which self-absorption could be examined.

Table 3.1 lists the alpha emitting isotopes used by other workers and includes ^{241}Am which was used for the work described in this thesis.

With the exception of natural uranium, all the isotopes have daughters with sufficiently long half lives that radiation from them will not contribute significantly to the total ionization produced.

TABLE 3.1 ALPHA EMITTING ISOTOPES USED FOR W VALUE MEASUREMENTS

AUTHOR(S)	ISOTOPE	HALF LIFE	PRINCIPAL	PRINCIPAL
			PEAK ALPHA ENERGY (MeV)	GAMMA ENERGY (keV)
Bortner and Hurst (1954)	^{239}Pu	$2.4 \times 10^4 \text{ y}$	5.14	13
Herwig and Miller (1954)	natural ^{238}U	$4.5 \times 10^9 \text{ y}$	4.19	48
Jesse (1952, 1953, 1955b, 1960, 1961, 1968a, 1968b)	^{210}Po	138.4d	5.30	unknown (<1%)
Bay and McLernon (1965)	"	"	"	"
Bay, Newman, and Seliger (1961)	"	"	"	"
Chappell and Sparrow (1967)	"	"	"	"
Harris (1971)	^{241}Am	458y	5.48	60

3.3 Some source mountings used by other workers

In order to reduce ambiguities relating to source energy, some workers have prepared very thin sources which emit into a 2π solid angle. Such a source prepared by Bay and McLernon (1965) is shown in figure 3.1.

Figure 3.1 Source mounting used by Bay and McLernon (1965)

^{210}Po is deposited on a collodion film which has received a thin gold film by evaporation. The presence of a conducting film is necessary to eliminate irregularities in field strength around the source. The thickness of the collodion films used was about $10\text{ }\mu\text{g cm}^{-2}$ but no value has been given for the thickness of the gold film. It is possible that some degree of backscattering took place in the gold film, resulting in a number of lower energy alpha particles being emitted.

Other workers, notably Bortner and Hurst (1954) have used more conventional planar sources with the isotope deposited upon a solid backing such as stainless steel. With such sources backscattered alpha particles may account for as much as 3% of the total number emission rate.

A collimated source as used by Bay and Newman (1961), shown in figure 3.2 is arranged so that alpha particles are emitted into the chamber through a thin collodion film which normally prevents ions produced within the collimator from entering the collecting volume.

Figure 3.2 Source mounting used by Bay and Newman (1961)

Their experiment was arranged to take account of the energy dependence of W by permitting only a limited alpha range in the chamber. The remaining alpha energy was spent in an aluminium target. A problem noted in their work demonstrates the difficulty of source preparation. Erroneous results were produced by dust on the collodion film and by slight damage to the film which allowed ions to be removed from the collimator housing by the collecting field.

3.4 Sources for the present work

3.4.1 Properties

In order to produce accurate reproducible results, the sources used in these experiments were required to meet the following specifications:

- (i) isotopic stability
- (ii) no tendency to creep
- (iii) mechanical rigidity and stability
- (iv) no reaction with heating to 200°C under vacuum
- (v) suitable geometry of emission

An eminently suitable isotope is ^{241}Am which decays with a half life of about 462 years to ^{237}Np , emitting alpha and gamma radiation. The use of an isotope with a half life of a few hundred years renders decay corrections virtually negligible, but does not require a very thick source, with consequent self absorption, to yield the required activity. A simplified decay scheme for ^{241}Am is shown in figure 3.3.

Figure 3.3 Simplified decay scheme for ^{241}Am (Lederer et al., 1968)

3.4.2 Commercially produced sources using ^{241}Am

The preparation of alpha emitting sources was not to be undertaken in view of the expertise available at the Radiochemical Centre at Amersham. Instead, it was decided to purchase standard planar sources. Three of these types are illustrated in figure 3.4 which shows an electrodeposited source, a vacuum sublimed source, and a piece of foil mounted in an aluminium holder.

Figure 3.4 Commercially produced ^{241}Am sources

Electrodeposited sources were, until comparatively recently, the best commercially available sources with little self absorption. Such sources were calibrated by the Radiochemical Centre. The technique of vacuum sublimation has now produced sources capable of better than 13 keV F.W.H.M. resolution. Americium foils are produced by embedding ^{241}Am in a silver matrix which is deposited upon a silver backing foil. A coating of silver or gold is then deposited on the silver matrix, effectively producing a sealed source. Such sources exhibit comparatively poor energy resolution but are easily and cheaply obtained. For high accuracy work at atmospheric pressure it was decided to use vacuum sublimed sources, while for later work at reduced pressure involving reduced energy alpha particles the cheaper

americium foils were used.

A typical energy spectrum measurement for a vacuum sublimed source is shown in figure 3.5. This was obtained using an Ortec grade A surface barrier detector with resolution better than 20 keV at 5.5 MeV. Electrodeposited sources show a greater self absorption, while a typical spectrum from a foil exhibited a F.W.H.M. resolution exceeding 500 keV with a distinct low energy tail.

Figure 3.5 Alpha particle energy spectrum from a vacuum sublimed source using a surface barrier detector

3.4.3 High resolution sources

It was considered necessary to use the best possible high resolution source (vacuum sublimed) in order to provide good calibration facilities for the energy spectrometer. The emission characteristics of the full energy sources used by other workers for investigations at relatively high pressures in which the full alpha particle range is spent in the gas were most closely approached by such a source.

most closely approached by such a source. Bortner and Hurst (1954), who used a similar planar source, determined W for helium some 4% higher than the previously accepted value.

The line broadening exhibited by a vacuum sublimed source at normal incidence from the detector is typically observed to be about 17 keV F.W.H.M. using a premium grade silicon surface barrier detector. Vacuum sublimed sources normally maintain their resolution for long periods.

A careful investigation showed the importance of backscattered alpha particles from the thick source backing. These alphas suffer energy losses in the scattering events and are consequently emitted with a reduced energy. It is possible that Bortner and Hurst, who used a 2π proportional counter to determine the total energy emitted into the gas, did not fully allow for the importance of this effect. It is proposed to extend this work by examining a number of sources prepared on different backing materials with varying degrees of polish.

3.4.4 Reduced energy sources

3.4.4.1 Introduction

In order that low pressure experiments could be carried out, it was necessary to reduce the energy of natural alpha particles emitted from ^{241}Am to ensure that their total energy was spent in the gas. The experimental limits and methods used will be discussed in the following sections.

3.4.4.2 Maximum permitted alpha energy

The design of the ionization chamber permitted a maximum overall range of 15 cm, while the proposed lower limit of the pressure range to be investigated in helium was 10 torr. Application of the Bragg-Kleeman Rule which states that the effective atomic stopping power is approximately proportional to the root of the atomic number, showed that the maximum permissible alpha particle energy in helium at the lower limit of the pressure range to be investigated was about 40 keV. An extension was later fitted to the ionization chamber giving a new maximum electrode separation

of about 45 cm.

The design requirement of energies of the order of 40 keV, at which capture and loss events are highly significant, precluded the use of a silicon surface barrier detector for energy measurements in view of the energy losses in the dead layer. A further difficulty is the possibility of non-linearity of W in silicon although Ewing (1962) has shown a linear response for hydrogen ions in the energy range 75-250 keV.

When reduced energy alpha particles are produced in the usual way by allowing natural alphas to penetrate an absorber, there are a large number of particles emitted with energies considerably less than the mean. This problem is particularly acute at low emergent angles. Given that suitable collimation could limit the angle of emission of particles to those approximately normal to the plane of the source, it is by no means certain that this would overcome the natural straggling in an absorber. A further research project under the direction of the author at the University of Surrey is currently showing progress in the investigation of energy straggling in absorbers and the significance of non-uniformity of thin solid films.

3.4.4.3 Methods available for producing reduced energy alpha particles

Several types of filter have been used to reduce the natural alpha particle energy available from isotopes. They include aluminium foils, mica sheets, mylar films, and variable pressure gas attenuators. Table 3.2 shows the workers who used these methods.

TABLE 3.2 METHODS OF ENERGY REDUCTION OF NATURAL ALPHA PARTICLES

METHOD USED	AUTHOR(S)
variable gas pressure in collimator	Bay and Pearlstein (1963)
" " " " "	Chappell and Sparrow (1967)
plastic film	Wingate, Gross, and Failla (1957)
mica	Jesse and Sadauskis (1955)
"	Jesse (1960, 1961)
evaporated gold film	Bay and Seliger (1960)

In an investigation of ionization produced in air and argon by single alpha particles as a function of their energy, Jesse (1950) used the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ and ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$ reactions to produce alpha particles with energies of 2.060 MeV and 1.474 MeV respectively.

The advantage of a mechanical filter such as aluminium or mica is that it can be readily replaced by another while the source remains in its original condition. Plastic films would not withstand the heat treatment that the outgassing of the present experiment demands, and such films tend to suffer from radiation damage and become porous. Some suffer from natural porosity and experimenters have often had to prepare several sources and select one which does not suffer from this defect. An additional and very important disadvantage is shown by the work of Bay and Seliger (1960) who showed that mylar films tend to lift off from the surface. In variable pressure experiments this makes a difference to the residual range of the alpha particle and is a possible explanation of the Wingate, Gross, and Failla work (1957), in which a field independent, but pressure dependent, recombination factor was observed.

There are then inherent advantages in vacuum evaporated films because they are permanent and inert. For these reasons an evaporated film was chosen as the most suitable method for energy reduction. The choice of material for evaporation was made after considering the ease of evaporation, the thickness required, and any possible reaction with the source. Although other materials were available, gold was chosen, a maximum thickness of about 10 μ being required. In repeated evaporations, there would be no problem with oxide formation as there might have been with aluminium.

3.4.4.4 Gold film evaporation procedure

A standard vacuum evaporation technique was used to deposit gold upon the surface of the source. The equipment used for this purpose was an Edwards High Vacuum Limited 6E4 evaporation unit. Figure 3.6 shows the evaporation chamber with a molybdenum boat containing gold wire, and a source in position ready for evaporation. The glass bell jar is seen in the background and a spare molybdenum boat in the foreground. An implosion guard was fitted over the bell jar during evacuation.

Figure 3.6 Vacuum evaporation chamber

The chamber was initially evacuated and the work subjected to a period of ionic bombardment. Evaporation took place at a pressure below 10^{-6} torr. It was found that if evaporation took place too rapidly, a grossly uneven deposit was formed upon the source. The boat-source spacing was usually about 5 cm, which represented the minimum distance for satisfactorily even coatings as observed by visual inspection.

A thickness monitor was not used during evaporation, since energy measurements were to be used for determining the energy reduction obtained. The microscopic ^{non}uniformity of the source coating was not considered to be significant.

3.4.4.5 Ionization experiments using reduced energy sources

Figure 3.7 shows some results for a source activity of about 3 μCi activity following successive evaporations of gold. The ionization current observed in argon decreases as expected with increasing gold thickness, but a pressure dependence was observed even at the lowest energies until pressures of a few hundred torr were reached. Examination of these sources

Figure 3.7 Ionization observed as a function of pressure and gold thickness

with the energy spectrometer revealed that a considerable number of particles were emitted with energies exceeding that required for total stopping in the gas.

In spite of careful attention to technique, the problems of producing a source emitting alpha particles of about 40 keV are considerable. In particular, at this energy, capture and loss is very significant and the implications of carrying out energy per ion pair measurements are clear; not only will there be significant variations due to particle type, but also to particle energy. The difficulty of determining the energy spectrum emitted from such a source is also apparent. Although the surface barrier detector is capable of linear response to low energies, the energy loss in the window itself is of the order of 20 keV. The use of a proportional detector to determine energy spectra will be of limited value having regard to the energy dependence of the energy per ion pair. The only realistic method for examining this region appears to be by using an accelerator capable of producing monoenergetic low energy ions.

3.5 Straggling of alpha particles in solid absorbers

The concern of the present work with reduced energy sources has led to a thorough investigation, under the direction of the author, of the energy straggling of alpha particles in solids. Using a vacuum sublimed ^{241}Am source and a silicon surface barrier detector, it has been shown that a region of decreasing straggling occurs towards the end of the alpha particle range. This effect, not previously reported in solids, has been related to stopping power data for low energies. Results for aluminium absorbers have already been reported (Sykes and Harris, 1971); the paper is included as appendix 3 to this thesis. Application of the technique to other solids has demonstrated the effect of non-uniformity of absorbers which are manufactured in different ways.

The use of energetic particles other than alpha particles is obviously attractive and fission fragments from ^{252}Cf have been examined. The significance of energy defect processes within the detector do not at present allow an entirely satisfactory conclusion regarding the decrease in straggling expected from stopping power considerations.

3.6 Preparations for experiments using the 2 MeV accelerator

With the acquisition of a 2 MeV Van de Graaff accelerator by the University of Surrey, a considerable programme of equipment reconstruction and improvement was carried out. The background of figure 3.8 shows the ionization chamber, now extended to allow a greater electrode separation, mounted on a trolley for easy access to the accelerator beam line.

Figure 3.8 The ionization chamber system at the accelerator laboratory

Larger vacuum pumps permit a more rapid pump down time. The analytical mass spectrometer with a complete and separate vacuum system is partially visible on the left of figure 3.8. Work is now nearing completion on the construction of an accelerator beam line, including stabilising slits, beam viewers, and collimators. Initial experiments will allow a beam of particles parallel to the electrodes to enter the ionization chamber through a thin window.

4.1 Introduction

In order to determine an absolute value of W it is necessary to know the total energy, E , being emitted into the gas. If an integration, or slow, measurement is to be made using alpha particles which are totally stopped within the gas, the total energy may be derived from the total number of alpha particles emitted into the gas together with their energy spectrum. However, other radiations from a radioactive source will also transfer energy to the gas. For ^{241}Am these may include heavy recoil ions, low energy gamma rays, and electrons. The significance of these additional emissions must be examined.

Initial experiments were designed to calibrate full energy sources, while later reduced energy sources were used.

Table 4.1 gives some W values determined by other workers and shows the calibration technique used in each case. The following general conclusions may be drawn from this data:

- (i) results obtained by different methods do not always agree
- (ii) Examination of the difference between fast and slow methods of ion collection in a gridded ionization chamber as carried out by Bay and McLernon indicates that both methods give the same result. This conclusion is given added weight by the results of Jesse who used fast ion collection in an ungridded chamber. It may be that electron collection on the grid was a factor in the work of Herwig and Miller which produced results somewhat higher than other methods for the gases studied.
- (iii) Agreement between Jesse and Bay, Newman, and Seliger is good although the source mounting was quite different in each case.
- (iv) It is difficult to reconcile the work of Bortner and Hurst with that of other workers since they did not apparently consider backscattered alpha particles from their platinum backed source. It may be that their method of source calibration in which a 2π proportional counter was used, failed to estimate correctly the relatively large number of backscattered alpha particles which reduce the mean alpha particle energy from the source.

Table 4.1 Some W values for five gases with different sources, calibration techniques and methods of ion collection

Author(s)	Isotope Used	Source Preparation and Mounting	Method of Source Calibration	Method of Ion Collection	W for Various Gases (ev per ion pair)		
					Air	Nitrogen	Carbon Dioxide
(Bay, Newman and Seliger, 1961)	^{210}Po	active deposit on thin gold film evaporated on to $10\mu\text{g}/\text{cm}^2$ collodion counter as check	2π counting in chamber with 2π methane proportional counter	slow (with simultaneous counting)	34.97 $\pm$ 0.07	36.38 $\pm$ 0.07	34.04 $\pm$ 0.1
(Bay and McLernon, 1965)	^{210}Po	"	"	"	-	36.02 $\pm$ 0.07	26.3 $\pm$ 0.07
		"	-	fast	-	36.33 $\pm$ 0.08	26.37 $\pm$ 0.07
(Jesse, 1960)	^{210}Po	active deposit on platinum	-	fast	34.96 $\pm$ 0.07	36.39 $\pm$ 0.07	-
(Bortner and Hurst, 1954)	^{239}Pu	electrodeposited on platinum	2π methane proportional counter	slow	35.0 $\pm$ 0.3	36.3 $\pm$ 0.4	34.3 $\pm$ 0.3
(Herwig and Miller, 1954)	natural uranium	$18\text{-}60\mu\text{g}/\text{cm}^2$ on brass	-	fast	-	38.8 $\pm$ 0.8	27.7 $\pm$ 0.6
(Chappell and Sparrow, 1967)	^{210}Po	electroplated on platinum	time of flight	slow (with simultaneous counting)	35.52 $\pm$ 0.20	36.65 $\pm$ 0.20	26.27 $\pm$ 0.15

The rigorous source calibration method to be described later was set up so as to enable us to allow for this effect.

(v) The recent work of Chappell and Sparrow, in which a time of flight method of source calibration was used, showed results for air and nitrogen that were higher than those previously accepted. No explanation has yet been put forward for this discrepancy, other than the possibility that results for air may depend upon the composition. Work carried out under the direction of the author has shown (Evans, 1970) that this effect could not have accounted for the observations of Chappell and Sparrow.

4.2 The energy spectrometer

4.2.1 Introduction

In order to measure the energy spectrum emitted from the sources, a grade A Ortec silicon surface barrier detector was mounted in a suitable chamber with provision for evacuation. The spectrometer will be described in greater detail in the following section.

Initially it was hoped to measure the source emission rate by standardising the apparatus with a reference source calibrated by the Radiochemical Centre. As will be shown later, the use of a standard reference source proved to be very inadequate.

With the use of reduced energy alpha particle sources, it was necessary to examine the spectrum emitted at various angles from the source. Energy calibration against a full energy high resolution vacuum sublimed source allowed unknown spectra to be measured.

4.2.2 The spectrometer

The spectrometer chamber was designed around the size of the sources chosen for the experiment and the source-detector distance likely to be of interest. With the electrode-electrode distance of 15 cm in the ionization chamber, a total length of about 30 cm was decided upon for the spectrometer chamber. The diameter of the chamber was chosen, partly empirically from mechanical considerations, and partly with a view to avoiding possible

scatter effects from the walls.

The spectrometer is shown in figure 4.1 and schematically together with the vacuum system and output connections in figure 4.2.

Figure 4.1 The energy spectrometer

Two facilities were designed to be incorporated in the chamber, both to be available under vacuum conditions. The first was a variable source-detector distance. For practical reasons, and in order to reduce input capacity, the detector was fixed in position in the lid of the chamber. Hence the source was made movable in a vertical direction, this movement being made with a sliding tube with double vacuum seals in the lid which could also be removed from the main body of the chamber. The source holder was made rotatable by an inner concentric tube running through the height adjuster. A rack and pinion method was used to transmit rotation of a calibrated knob on the top of the instrument to the central rod, a further

rack and pinion at the source holder re-translating vertical motion into rotation of the holder. Calibration of the knob permitted measurement of the source angle to within 7.5° . Some backlash in the gearing led to some uncertainty in this measurement and later modifications allowed selection of an angle to within 1° .

Figure 4.2 Schematic diagram of energy spectrometer system

The chamber was constructed from brass with mild steel endplates, these being chosen for reasons of economy consistent with mechanical strength. The two stage rotary pump evacuated the chamber to a pressure of 10^{-3} torr. Original experiments were carried out without a cold trap but the measured

resolution was observed to fall from about 20 keV F.W.H.M. at 5.5 MeV to greater than 50 keV. Since it was considered possible that the vacuum sublimed source and not the detector was the cause of this failure, it was returned to the Radiochemical Centre for examination. Measurements carried out (Byrne, 1969) indicated that possible contamination of the source surface had caused the deterioration. Further examination of the system revealed that the detector had also deteriorated, presumably due to poisoning of the device with hydrocarbon oil vapour. Since fitting a cooled pipeline trap in the vacuum system, no further difficulty has been experienced with another detector which has been in use for over a year.

4.2.3 Losses associated with spectrometer measurements

Energy losses in the residual gas and dead layer of the detector were negligible at full alpha particle energy although losses in the latter would be of increasing significance at reduced energies. Count rate losses due to scatter from the residual gas are also negligible at the pressure used.

4.2.4 Use of a reference source to calibrate unknown emission rate

4.2.4.1 Procedure

A semicircular perspex holder was mounted in the spectrometer to enable a reference source or a blank area to be rotated to face the detector. The source whose emission rate was to be measured was mounted parallel to the detector and alpha particles were counted when the perspex holder was rotated to allow their passage. The background count rate, determined by rotating the blank perspex to face the detector, was observed to be negligible.

The count rate observed from the reference source allowed the geometrical efficiency of the system to be calculated and hence the 2π count rate for the unknown source to be determined.

As a check on this result the emission rate was also estimated from

a measurement of the ionization current produced in nitrogen, assuming the well established value of W for nitrogen as 36.6 eV per ion pair. The results are shown in table 4.2.

TABLE 4.2 COMPARISON OF METHODS USED FOR MEASUREMENT OF SOURCE EMISSION RATE

METHOD USED	OBSERVED EMISSION RATE (c.p.s.)
comparison with reference source	243
ionization current	234

The conclusion drawn from this work was that the reference source was probably in error. A new calibration certificate revealed that the emission rate was about 10% below that originally recorded and, in spite of careful handling, loose activity was present on the surface. The new source calibration figure, however, gave an expected emission rate for the unknown source of 218 c.p.s., some 6% lower than that inferred by ionization measurements.

4.2.4.2 Discussion

The difficulties and errors arising from the use of a reference source method of calibration of unknown source emission rates will be discussed in this section.

(i) Calibration of reference source. Although carried out by a reputable laboratory using a 2π proportional counter, there was no guarantee that the figure established would remain valid for a period of time. There is no reason to suppose that anything other than gross damage would change the emission rate, but, nevertheless, this appears to have happened in the early life of the source. Possibly this was a result of poor adhesion of the radioactive material to the backing support.

(ii) Accuracy of calibration. The Radiochemical Centre emission rate was quoted to be within 3%, a figure which includes three standard deviations from the observed count rate and a systematic error. Although the overall accuracy has now been increased to 2%, it is clearly unsatisfactory.

With longer counting times the accuracy of measurement in a 2π gas counter may be increased considerably. The National Bureau of Standards laboratory normally calibrates to within 0.1% for emission rate.

(iii) Backscattering of alpha particles. The phenomenon of alpha particle backscattering from a thick backing material has long been recognised. Particles scattered in this way emerge at angles within about 15° to the plane of the source. Radiochemical Centre calibrations assume that not more than 3% of the total 2π emitted count rate will be due to this effect. It has been shown (Walker, 1965) that the magnitude of the backscatter effect is a function of the atomic number of the backing material. Since the degree of polish of the surface is also an important factor, as are the energy losses due to scatter events, it is necessary to examine each source particularly with reference to alpha particles emergent at low angles near the plane of the source. If sources are to be used in a low geometry configuration, this problem may be avoided.

4.2.5 Energy emission measurements using the spectrometer

A programme of work was undertaken to determine total particle and energy emission from ^{241}Am sources over 2π . The results have already been reported (Yeates, Harris, and Doust, 1970) and the paper is included as appendix 2 to this thesis.

The solid angle geometry of the spectrometer was calculated and a number of energy spectra recorded at various angles of emission from the source. Each spectrum from the 400 channel analyser was corrected for different counting times and for the variation in the total solid angle of emission as a function of a given angle of emission. A computational summation of all particles emitted over 2π was carried out and a value for total energy emission over 2π derived from calibration of the multichannel analyser. Experiments were carried out using a vacuum sublimed and a gold coated foil source. Figures 3 and 4 in appendix 2 show corrected spectra obtained for each source at various angles of incidence, while figures 5 and 6 indicate the total number spectra over 2π . The alpha backscattering effect from the vacuum sublimed source is clearly shown in figure 7. Number emission rates calculated in this way were compared with observations using a 2π proportional counter. Excellent agreement was obtained for the gold coated source but relatively poor agreement for the vacuum sublimed source.

It was concluded that, for both sources, the number and energy of alpha particles emitted varied very greatly with the angle of emission.

4.3 The 2π proportional counter

In order to calibrate sources for energy per ion pair studies it was necessary to carry out accurate emission rate measurements over 2π . The use of a 2π proportional counter has already been mentioned in the previous section, the results showing the need for a check to be carried out against low geometry extrapolations.

A simple 2π proportional counter was constructed but it was soon recognised that, for high emission rate measurements, special counter characteristics would be required. The investigation resulted in a 2π counter capable of measurement up to at least 5×10^5 alphas per second (Yeates, Harris, Doust, and Parish, 1969). The publication is included as appendix 1 to this thesis. The factors limiting counting speed were carefully examined, particularly the mechanical and electrical design and the choice of counting gas.

4.4 Summary

A programme of research has been carried out with the aim of elucidating the number and energy emission rates of alpha particles over 2π from planar sources. Both vacuum sublimed and matrix foil sources have been examined, the latter being coated with gold to reduce the alpha particle energy. The significance of backscattered alpha particles has been shown by measurements in a low geometry spectrometer specially constructed for the work. Alpha particle emission rates have been carried out over 2π geometry, and a new gas flow proportional counter has been reported with count rate capabilities up to 2×10^5 alphas per second. It is suggested that previous workers have not fully considered the problem of source calibration when carrying out determinations of the energy per ion pair.

CHAPTER 5. THE IONIZATION CHAMBER

5.1 General design and construction

The design of the present plane parallel plate ionization chamber follows that of Bortner and Hurst (1954) although, since low pressure investigations were intended, the dimensions were increased. Table 5.1 compares the electrode spacing and diameters of the two chambers.

TABLE 5.1 DIMENSIONS OF IONIZATION CHAMBERS

	Bortner and Hurst (1954)	Harris (1971)
electrode dia. (cm)	19	45
electrode sep. (cm)	6.85	15
min. pressure for ^{241}Am alpha	200	100
absorption in helium (cm of Hg)

Figure 5.1 The ionization chamber

Figure 5.1 shows the construction of the ionization chamber. The large diameter of the electrodes relative to their separation leads to a uniform electric field within the collecting volume. An approximate minimum working pressure, at which the entire ^{241}Am alpha particle range is contained within helium, is shown in table 5.1. Thick backed alpha sources were mounted in the high potential electrode. An extension to the ionization chamber has since been fitted, allowing a maximum electrode separation of 45 cm.

Most of the experiments were carried out using the slow method of ion collection in which electrons, or negative ions, were measured as a current. External source emission rate and energy spectrum measurements were carried out using the energy spectrometer and 2π proportional counter already described. The later introduction of a Frisch grid within the main chamber permitted pulse collection of electrons. The energy resolution of this arrangement was limited by the capacitance of the collecting electrode with respect to earth.

The main body of the ionization chamber was constructed from stainless steel. The chamber lid and source entry port were both sealed with Viton "O" rings, which enabled the chamber to be heated to 200°C without failure of the seals. Additionally, Viton has a lower outgassing rate than rubber. These and some other materials are compared for outgassing in table 5.2.

TABLE 5.2 OUTGASSING OF MATERIALS AT ROOM TEMPERATURE (Dayton, 1961)

MATERIAL	OUTGASSING RATE (litre torr sec ⁻¹ cm ⁻²)
rubber	$2.2 \cdot 10^{-8}$
viton	$2.0 \cdot 10^{-8}$
P.T.F.E.	$2.5 \cdot 10^{-8}$
stainless steel (baked)	$5 \cdot 10^{-15}$
stainless steel (unbaked)	10^{-7}

The collecting electrode was mounted on a vacuum leadthrough using quartz as the insulator and Viton "O" rings as seals.

The high voltage electrode was made negative with respect to the collecting electrode which was virtually at earth potential. Although in some geometrical configurations this arrangement is disadvantageous for satisfactory ion collection, the use of an earthy collector eliminates difficulties of high voltage blocking capacitors and increased collector capacity.

The high voltage electrode was supported from the lid of the chamber by means of glass fibre reinforced P.T.F.E. bolts. Investigations showed that the presence of glass fibre reinforcement did not detract from the insulating properties of the bolts. The electrical lead-through for the high voltage supply was similar to that used for the collecting electrode, but this was later replaced by a coaxial version with ceramic insulation, which allowed an additional high voltage supply for a Frisch grid to be admitted to the chamber.

To allow outgassing of the clean and polished stainless steel, and to enable it to be used at elevated temperatures if required, electrical heaters were fitted to the outer surfaces of the chamber. These permitted baking to be carried out to 200°C, this limit being set by the Viton seals. Thermocouples were placed on the surface at several points and the temperature was monitored by a six channel recorder, a switch allowing selection of further thermocouples monitoring the gas purification plant. A cut-out was provided to prevent overheating. Mineral wool insulation aided even temperature distribution and economy of heater loading.

Stainless steel diaphragm valves were fitted to the lid and base of the chamber, providing facilities for gas entry and circulation. The presence of small quantities of chemicals from the purification plant at various times caused these valves to lose their leak-tight integrity and they were later replaced with standard Viton diaphragm valves.

As the intention was to study gases at varying pressures, it was necessary to know the gas pressure within the ionization chamber. It was decided that a system by which a known mass of gas was introduced into the system would be desirable, having due regard to experiments involving the addition of known quantities of impurities. Accordingly a

'shared volume' system, shown in simplified form in figure 5.2, was designed, in which the supply cylinder filled a gas sampling bomb 'V' to a known and measured pressure at room temperature after allowing for thermal equilibration, the valve being closed.

Figure 5.2 The 'shared volume' system

At this stage, the ionization chamber and gas purification system was undergoing evacuation. With the vacuum system closed off, the valve was then opened and the gas in 'V' admitted to the main system. The pressure fell in the ratio of volume 'V' (including associated pipework) to the volume of the ionization chamber (and gas purification apparatus). The volumes of various components of the system were calculated from their dimensions, from which it was determined that the volume of the ionization chamber (including the gas purification system) was 53.1 litres, and the volume of the gas entry system was 0.5 litre.

The choice of pressure measuring instrument was guided by criteria of versatility, reliability, accuracy, and the extent to which the device may introduce unwanted contamination. Aneroid and Bourdon gauge instruments with direct reading scales and an accuracy of at least 1% have been used for many years in similar environments and are well proved. However, it was decided that a strain gauge pressure transducer would offer advantages over the more conventional gauges and an instrument covering the range of 0 - 100 pounds per square inch was supplied by Consolidated Electrodynamics together with a control amplifier and suitably calibrated meter. The instrument was calibrated at zero pressure (10^{-2} torr as measured on a

Pirani gauge) and atmospheric pressure as measured with a Fortin barometer. Linearity and hysteresis characteristics of the system allowed pressure readings to be made to within 0.5% of the absolute value. A digital voltmeter was coupled to the output of the amplifier and allowed accurate observations.

As a result of discussions with the British Oxygen Company, small bore oil-free copper tubing was fitted to the gas entry system between the supply cylinder and the sampling bomb. A flushing procedure was adopted before admitting gas to the sampling bomb.

5.2 High voltage supply

With the need to measure ionization currents of less than 10^{-9} amps at an accuracy of within 0.1% by the charge integration method, the high voltage supply was required to be stable to within a few parts per million during the period of charge integration. An R-C smoothing circuit was constructed in order to improve the characteristics of the available supply unit, an additional problem being breakthrough of the high frequency oscillator used in the generation circuit. Time constants of the order of 800 seconds were necessary to achieve the best stability for the lowest measured currents. An automatic switching system was constructed, using cam controlled microswitches, in order to perform measurements after the long period required for equilibration of the output voltage. At low voltages (up to 600 volts), dry batteries with suitable safety resistors were used, although the stability of these with temperature variations is not particularly good (about 0.01% per $^{\circ}\text{C}$).

5.3 Frisch grid

In non-electronegative gases, electrons resulting from the passage of an ionizing particle may be collected within a few microseconds. The slower positive ions exert an induced charge effect which may be screened from the collecting electrode by means of a grid (Bunemann, 1949). Fast counting of alpha particles in air has also been carried out (Bay, 1960), but with poor energy resolution.

For the purposes of the present work, the advantages of using a Frisch grid were considered to be the ease of 2π counting, and the possibility of investigating possible differences between fast and slow methods of ion collection. Using the theory developed by Bunemann (1949), suitable parameters were calculated from which a grid was constructed. Stainless steel wires of 0.122 mm diameter were spaced with a pitch of 2 mm by means of locating pins silver soldered into a square section of stainless steel, which had been rolled into a diameter of about 45 cm. Some Frisch grid parameters for this and other workers' grids are shown in table 5.3.

TABLE 5.3 SOME FRISCH GRID PARAMETERS

AUTHOR(S)	WIRE RADIUS (mm)	WIRE PITCH (mm)	GRID-COLLECTOR SEPARATION (cm)
Herwig and Miller (1955)	0.04	2	1.25
D'Orval (1960)	0.04	1	1.2
Bay and Pearlstein (1963)	0.041	0.82	1
Harris (1971)	0.061	2	1
"	"	"	5

Figure 5.3 shows the grid together with the high voltage electrode and the coaxial lead-through. P.T.F.E. supporting pillars, bolted to the high voltage electrode and the grid, held the latter in position.

Figure 5.3 The high voltage electrode and Frisch grid

CHAPTER 6. THE VACUUM SYSTEM

6.1 Design considerations

The primary objective of the vacuum system was that it should be capable of evacuating the ionization chamber to a pressure at which the effect of the residual gas would be negligible in the presence of the specimen gas. It was considered that a conventional system capable of an ultimate pressure of about 10^{-7} torr would satisfy this condition.

The optimum pumping speed was considered to be set by a balance between the cost of equipment and a reasonable pump down time. Calculations allowing for the outgassing of stainless steel showed that the time to reduce the pressure to 10^{-6} torr would be approximately 14 hours using a 50 litre minute⁻¹ diffusion pump. Initially backstreaming from the diffusion pump to the ionization chamber was limited by a thermoelectrically cooled baffle, but later examination of the residual gases using an analytical mass spectrometer showed a high concentration of organic vapour, and accordingly a later design of the vacuum system included a liquid nitrogen cooled baffle to eliminate this problem.

With the complete system in use for helium with purification traps and chemicals, the actual pump down time was at least 14 hours and usually considerably longer. This difficulty arose primarily through the greatly increased outgassing rate of the purification chemicals and, to a lesser extent, the restricted pumping speeds around the pipework.

6.2 The pumping system

The conventional high vacuum system used to evacuate the ionization chamber and gas system is shown in figure 6.1. Two 35 litre minute⁻¹ rotary pumps were used, one to rough pump the entire system, and the other as a backing pump to the 2" oil diffusion pump. Silicon oil was originally chosen for use in the diffusion pump, but later work with mass spectrometer leak detectors and analysers required a change to apiezon oil, to avoid the tendency of silicon oil to form insulating deposits on the electrode

systems. Since the main chamber was to be heated to 200°C, a water cooled manifold was placed between the chamber and the diffusion pump.

Also shown in figure 6.1 is a secondary high vacuum system, employing a 1" oil diffusion pump, which was used as a means of preventing inleaks around the high voltage and collecting electrode feed-throughs and the source entry port.

Figure 6.1 The high vacuum system

6.3 Pressure measurement

Pressure measurements were made throughout the system by Pirani gauges in the range $1 - 10^{-3}$ torr and Penning gauges in the range $10^{-3} - 10^{-6}$ torr. An ionization gauge would have been more satisfactory at the lower pressure but was not available during the course of this work. Residual gas analysis of the vacuum was carried out when a mass spectrometer became available.

6.4 Leak testing

With the size and complexity of the vacuum system it was found essential to provide a range of leak testing instruments. These are summarised in table 6.1.

TABLE 6.1 LEAK TESTING INSTRUMENTS

INSTRUMENT	USEFUL PRESSURE RANGE	SENSITIVITY
discharge tubes	atmos. - few torr	gross leaks
halogen detector	0.5 torr	10^{-5} litre torr sec ⁻¹
mass spectrometer	10^{-4} torr	10^{-12} torr (min. detectable partial pressure

6.5 An investigation of leak tight integrity and outgassing rate of the apparatus

6.5.1 Introduction

The extent to which true leaks may be detected in a complex system is limited by the number and nature of the manifolds and unions. When a pressure of 10^{-6} torr had been reached, as measured with a Penning gauge, it was frequently found that further leak testing with the mass spectrometer leak detector yielded little result, i.e. no individual leaks exceeding about 10^{-11} litre torr second⁻¹ could be detected. Accordingly it was decided to rely upon long term pressure rise measurements following isolation of the evacuated system to indicate the extent of true leaks and to examine the outgassing rate. A series of experiments was carried out to investigate, for example, the significance of Penning gauge siting within the vacuum system as these gauges were used to make the observations of pressure rise, and the effectiveness of the secondary vacuum system which was designed to reduce inleaks at the electrode feedthroughs and the source entry port. Outgassing measurements were made to check the performance of the system with purification chemicals in the traps. Before an experimental run in a gas was commenced, a pressure rise test was carried out to test the overall integrity of the apparatus.

Two Penning gauges were used in these experiments, the first being sited at the bottom of the ionization chamber in the high vacuum pumping line near the diffusion pump, and the second at the top of the ionization chamber in the gas entry system. As a result of pumping constrictions within the system, the upper gauge normally read 10^{-5} torr while the lower read 10^{-6} torr. Pressure rise data was initially collected using both gauges, but later routine measurements exclusively used the lower gauge which was permanently installed as part of the vacuum system. The gauge control units were modified to allow a chart recorder to be used to provide a permanent record.

6.5.2 Results

No significant difference was obvious between measurements made with alternative arrangements of the vacuum system, although a decrease in total apparent leak rate with increasing pressure was observed and this was taken to indicate that outgassing was a significant contributory factor. A typical measured value for the total apparent leak rate was 1.7×10^{-6} litre torr sec.⁻¹ as measured at a mean pressure of 5×10^{-5} torr, reducing to 0.9×10^{-6} litre torr sec.⁻¹ at 3.5×10^{-4} torr. Following a bakeout of the system at 200°C for at least 72 hours, a reduction by a factor of two or three was usually observed although prolonged baking often led to spontaneous inleaks arising, probably due to poorly seated seals.

Following the introduction of purification chemicals, a greatly increased outgassing rate was observed but, following prolonged baking of the complete system at 200°C a total apparent leak rate equalling that of the vacuum system alone was achieved.

6.5.3 Summary

This series of experiments was carried out primarily to investigate the leak tight integrity of the complete system. To a limited degree of accuracy in view of the poor instrumentation used, it allows assumptions regarding the probable rate of outgassing of the apparatus although this is only satisfactorily carried out if the relative concentrations of the residual gases are measured as a function of time.

It has been demonstrated that the leak tight integrity of the system was of a good high vacuum standard and that the overall apparent leak rate in a thoroughly baked system was adequate for work to be carried out in pure helium at atmospheric pressure.

CHAPTER 7. GENERAL CONSIDERATIONS OF THE EXPERIMENTAL MEASUREMENT OF THE ENERGY PER ION PAIR IN HELIUM

7.1 Introduction

It is the purpose of this chapter to consider some general topics of significance in the measurement of the energy per ion pair in helium. Initially a discussion of some factors influencing the real and apparent ion yield is presented, followed by a description of the design and operating characteristics of the gas purification plant. The current measurement system is then described, together with an automated technique that was developed to allow periods of unattended operation. The significance of leakage and background measurements is also discussed.

7.2 Factors influencing ionization produced and collected

In any total ionization experiment it is necessary to ensure that the observed ionization yield corresponds to that actually produced in the gas by irradiation. The particular unrelated problem of secondary effects due to contaminants in a gas has already been discussed, as has been the effect of molecular ion formation in pure helium, but there remains a number of factors which are capable of increasing or decreasing the observed ion yield, and these will now be considered.

An increase in ion yield may arise through secondary ionization resulting from acceleration of the primary electrons, although this is energetically impossible in view of the relatively low field strengths and gas pressures used in the experiments described in this thesis. An electrical discharge within the apparatus is less unlikely and did actually occur during early testing. The observed ionization current is extremely unstable and permits rapid diagnosis of this condition. In an exhaustive treatment of possible sources of ionization it could perhaps be mentioned that thermionic emission from the walls of the apparatus is negligible, certainly at the temperatures used, and photoelectric processes as a result of the approach of positive ions to the cathode are also negligible in current ionization chambers.

An effective loss of ion yield in ionization chambers arises primarily as a result of recombination of ion pairs. Two general categories of recombination have been recognised. Initial recombination occurs within individual particle tracks and depends upon the L.E.T. of the ionizing particle and upon the strength of the collecting field and its direction in relation to the particle track. General, or volume, recombination occurs when ions from a particle track recombine with those from another track. Both categories have received considerable attention in the literature and Niatel (1967) discusses them in his study of recombination in a parallel-plate ionization chamber. It is clear that volume recombination will be of little significance under the modest conditions of ion density and gas pressure used in the present work, particularly since non-electronegative gases are of primary concern. Of greater significance is initial recombination, with the high linear ion density arising from alpha particle tracks. A correction is usually derived by extrapolating the observed ion yield to infinite collecting fields (Wyckoff and Attix, 1957). Although considerable studies have been carried out into recombination difficulties, no better or more convenient technique has been demonstrated. It may be shown theoretically that the most satisfactory technique of reducing initial recombination is to align the collecting field normally to the particle track, but there is no evidence to show that any effective improvement is obtained by adopting this method. Assuming that all the irradiation particles are completely stopped within the gas before reaching the walls or other constructions within the apparatus, it is evident that ions may be effectively lost by failure to be collected on the appropriate electrode. The former assumption may be justified by appropriate selection of gas pressure so that observation of the ion yield will indicate total absorption. Gas pressures were chosen by this method for the present ionization chamber, the electrode spacing and diameters being chosen to eliminate the second problem by providing a uniform field strength within the collecting volume. The effect of backscattering from the gas to the supporting electrode system will be negligible in view of the arrangement adopted.

7.3 Design and operating parameters of the gas purification plant

7.3.1 Introduction

The need to maintain an extremely high degree of helium purity has already been indicated through the work of Jesse (1952), although the tolerable limit for impurities is uncertain in view of his rather low value for the energy per ion pair in helium. Commercially available helium may be obtained either in a spectroscopically pure grade or in a normal commercial grade containing a few parts per million of several contaminants. The cost of spectroscopically pure gas is prohibitive, particularly in the quantities required, in addition to which the maintenance of purity would be unattainable. With the selection of commercial grade helium a gas purification plant was necessary to remove the contaminants from the supply gas, and to maintain a high degree of purity by removing impurities desorbed from the walls of the apparatus during the course of an experiment.

Early workers in the field have used either chemical or physical methods of purification, and frequently a mixture of both. The most satisfactory method is probably that using liquid helium to trap all impurities. It has been suggested to the author by Bortner (1968) that heated purified uranium chips, which are extremely reactive, may be a suitably efficient trap. Both these methods are expensive and, in the latter case, hazardous. In recent times the need for a continuous supply of ultra pure helium has been amplified by the development of gas cooled reactors in which the coolant is helium and where the presence of unwanted contaminants leads to a poisoning of the core.

In previous energy per ion pair measurements no attempt has been made to assess the likely purity of the helium used, and it was necessary to consider possible ways of carrying this out in the present work.

7.3.2 Physical purification

Some workers have claimed to produce pure helium by passing the gas through a cooled sorption trap, usually activated charcoal prepared by heat treatment on dense wood charcoal which is heated under vacuum to remove occluded gases. The term sorption describes both adsorption, in which molecules condense on a surface, and absorption, in which molecules enter a porous material and are trapped. Both processes may be contrasted with chemical reactions in which the molecule actively combines with the

trapping reagent. Activated charcoal has a high sorption value which increases considerably if the temperature is reduced, as may be seen in table 7.1.

TABLE 7.1 SORPTION TRAPPING USING ACTIVATED CHARCOAL (Spinks, 1963)

GAS	VOL. AT N.T.P. OF ABSORBED GAS PER ML OF CHARCOAL(ml)	
	0°C	- 187°C
oxygen	18	230
nitrogen	15	155
hydrogen	4	135
helium	2	15

In addition to active sorption on a substrate, a cooled trap will be expected to remove all low boiling point gases, particularly organic vapours. Liquid nitrogen or air were the coolants used in the course of the experiments.

Activated charcoal was later abandoned in favour of molecular sieves (crystalline metal alumino-silicates) supplied by Union Carbide Ltd. Cations exposed in the crystal lattice of these substances are responsible for the selective adsorption which is shown for polar molecules. The surface area of the sieve is available as an absorbing site for all molecules. A number of sieve types are shown in table 7.2 which indicates the diameter of molecules which are excluded from the uniformly porous crystalline structure. Also shown is an indication of the saturation water capacity, the two figures being the limits for varying pellet sizes of the sieve. By reducing the temperature of the absorbant, a lower water vapour pressure will result, as indicated in isotherm data supplied by the manufacturers.

TABLE 7.2 MOLECULAR SIEVE TYPES (Union Carbide U.K. Ltd.)

SIEVE TYPE	EQUILIBRIUM WATER CAPACITY (% weight)	MOLECULES EXCLUDED
3A	23 - 20	3A°
4A	28.5 - 22	4A°
5A	28 - 21.5	5A°
10X	36 - 28	8A°
13X	36 - 28.5	10A°

7.3.3 Chemical purification

Chemical purification is normally used as a necessary addition to efficient physical purification provided by a cooled sorption trap. Early experiments used heated calcium chips to remove nitrogen, oxygen, and hydrogen although this has been shown (Gibbs, 1956) to be relatively inefficient for nitrogen, particularly at the temperatures used (usually about 400°C). In an attempt to reproduce the conditions of previous workers, calcium chips were used in the early stages of the present work.

A more sophisticated helium purification train for use with gas cooled reactors has been designed by Berry (1962). This system used titanium chips at 800°C to remove nitrogen, and hopcalite (a mixture of copper and manganese and their oxides) at 400°C to trap oxygen as copper oxide, and to oxidise hydrogen to water and carbon monoxide to carbon dioxide. Later work by Bourke (1965) showed that the titanium trap was unnecessary if cooled molecular sieve was used to trap nitrogen. In view of this, and to avoid the difficulties associated with high temperature systems, a titanium trap was not used in the present work. Instead, a hopcalite trap, designed to operate at 400°C, was fitted to the system.

7.3.4 Design of purification and gas entry systems

The purification traps and pipework were constructed of stainless steel, using a commercial coupling system to ensure that the arrangement could easily be changed for specific needs. Viton "O" rings were used throughout. Electrical bakeout tapes were fitted to all pipework to permit outgassing and were designed to operate at 200°C, the temperature being monitored by a multichannel pen recorder. The hot trap was fitted with a heating ^{tape} ~~trap~~ designed to operate at 400°C, brass cooling fins at the extremities of the trap reducing heat conduction along the pipework. A temperature controller was fitted to this trap.

Recirculation of the gas was carried out by means of a diaphragm pump, specially modified to ensure that only Viton or stainless steel was in direct contact with the gas. The normal gas circulation rate around the purification system was measured using a calibrated flowmeter and found to be about 1 litre per minute at normal operating pressure.

The gas circulation system is shown diagrammatically in figure 7.1 which also shows the gas entry arrangement from the supply cylinder. The ionization chamber together with part of the gas purification system is shown in figure 7.2.

Figure 7.1 Gas circulation system

Figure 7.2 The ionization chamber and gas purification system

Siting of the purification traps was not considered to be important although initially it was decided to position the hot trap beneath the ionization chamber so that convection currents would aid gas circulation.

Indeed, the work of Bortner and Hurst (1954) included a hot calcium trap only, placed in a similar position, convection currents being the sole form of gas recirculation. In the present work gas from the top of the chamber was passed through a cold trap, followed by the recirculation pump. The inlet and outlet positions on the ionization chamber were arranged to ensure mixing of the gas within the chamber. Although several rearrangements were made of the trap positions and direction of flow, the purity of the helium produced was remarkably constant, as measured by the observed ionization current.

Figure 7.3 shows the recirculation pump, the cold trap fitted with an electrical bakeout tape, and the hot trap.

Figure 7.3 The gas purification system

7.4 Ionization current measurement

7.4.1 The measuring system

The basis of the measuring system was a conventional vibrating reed electrometer (Vibron 62A manufactured by Electronic Instruments Limited). The converter unit of the instrument was connected to the ionization chamber by a short length of P.T.F.E. insulated, non-microphonic wire, and the electrometer was capable of measuring currents somewhat lower than 10^{-16} amps. Although difficulties arose initially from the influence of stray currents, these were eliminated by careful attention to earth loops and screening. The significance of filtering on the high voltage supply has already been pointed out.

The usual function of electrometers is as high gain amplifiers, with negative feedback applied to the input so that the input potential is virtually zero. This method approaches the more basic circuit developed by Farmer (1955), in which the electrometer is maintained at a null by means of a variable potential supply of high stability. Similar techniques have frequently been used (Lea, 1937; Kemp, 1954) but have now been superseded by the high open loop gain electrometers currently in use.

In order to avoid the uncertainties involved with the use of high value resistors, the more usual method of low current measurement was chosen, namely, the integration of accumulated charge on a capacitor. The time of charge collection, together with the potential appearing across the capacitor at the commencement and completion of charge collection, and a knowledge of the capacitor, provided the data by which the mean ionization current could be calculated. The present measurements allowed charge to be integrated across the total input capacity including that of the ionization chamber and the electrometer, the latter consisting of the input circuitry proper and a plug-in capacitor on the converter unit. The plug-in capacitors used were high stability polystyrene types, frequently checked against a standard bridge. By using alternative plug-in capacitors, the effective input capacitance could be determined for absolute measurement although this formed only an insignificant part of the work, in view of the importance of measurements relative to other gases in which stability was the prime concern. A highly stable potential supply, itself calibrated

against a standard cell, was available to calibrate the electrometer, the output of which could be connected to a digital voltmeter. A stopwatch was used for timing initially, but was replaced by timing pulses derived from the electricity mains and, later by a crystal oscillator.

7.4.2 An automatic system for current measurement

The desirability of providing a cheap automatic current measuring system was obvious and an original technique for this was developed, the essential features of which are shown in figure 7.4.

Figure 7.4 Automatic current measuring system

The principle of the technique is that two values of potential appearing across the integrating capacitor are arbitrarily chosen, and the time taken to charge the capacitor from the lower to the upper limit is measured by a suitable counter. For convenience the lower potential limit was chosen to be zero, while the upper limit was determined by a back-off potential available from the electrometer. A separate experiment was carried out to calibrate and determine the stability of this potential.

When the system is in operation a command pulse simultaneously opens the discharge switch 'S', and the gate, allowing ionization current to flow into the integrating capacitor and timing to commence. The second command pulse is derived from an optical galvanometer which is coupled to the electrometer. The light spot from the galvanometer traverses a photosensitive cell at a pre-determined excursion of the electrometer amplifier through zero.

Since charge integration times were never less than 100 seconds, electromechanical counters operating at 10 counts per second were used, in conjunction with a programming system which allowed experiments to be carried out automatically with only infrequent recording of the counter displays which provided the timing information. Timing pulses were derived initially from the mains frequency. The system was later refined by the replacement of the counters by a printing electromechanical device with facilities for automatic resetting. Control relays operated the start and stop functions of the system together with the print, reset, and restart functions.

The accurate detection (within a few millivolts) of zero output from the electrometer could be carried out by a d.c. amplifier coupled to a Schmitt trigger or similar device, although the additional circuitry required would not be inconsiderable in view of the sensitivity and very low drift required. Instead an optical galvanometer was used for this rather novel application, since such instruments have adequate sensitivity at the low currents and voltages involved, and are inherently stable. A difficulty remained in the linear sensitivity of the galvanometer which results in serious damage to the movement if overload is applied. Such a condition will arise at the commencement of a charge integration when a considerable fraction of the electrometer output (12 volts) may be applied across the galvanometer. This problem was overcome by the use of two semiconductor diodes in a back to back configuration in series with the galvanometer. By this means the instrument could be operated at maximum sensitivity with a greatly flattened response at potentials exceeding a few millivolts. Since the diodes have a finite resistance of the order of several thousand ohms at zero volts, the consequent reduction in sensitivity of the galvanometer can be offset by using a longer optical path.

7.4.3 Stability and errors

The stability of the electrometer was quoted by the manufacturers to be ± 0.1 mv in 12 hours. Frequent checking of the zero demonstrated that, after a long warm up period of over a day, the stability was well within this figure which represents an error of much less than 0.01% on all measurements. Long term drift was similarly demonstrated to be low, and was corrected by daily zeroing of the instrument.

The lower potential limit, nominally zero, was subject to the usual problems arising from charge generation associated with switch contacts. Since the maximum likely charge generated in this way was 10^{-15} coulombs, the phenomenon was negligible in ionization measurements with typical integrated charge values of $2 \cdot 10^{-10}$ coulombs. A later version of the measuring system eliminates this possible error.

The upper potential limit is determined by the stability of the electrometer and the internally derived back-off potential, the overall error being due to the latter. A contribution of $\pm 0.1\%$ was assigned to this quantity, errors in the optical lever system being negligible.

All electromechanical relays used for controlling the system were standard types with operating speeds of the order of milliseconds and no error could be attributed to them, but timing operations were carried out by reference to the public electricity supply frequency, and an upper error limit of $\pm 0.6\%$ could occur.

For the comparative ionization measurements, the current measuring system was considered to be stable to within about $\pm 0.7\%$.

CHAPTER 8. MEASUREMENT OF THE ENERGY PER ION PAIR IN HELIUM RELATIVE TO OTHER GASES

8.1 Introduction

If the absolute value for energy per ion pair is known for one gas then a relative value for another may be obtained by a simple measurement of the total ion yield for both under the same irradiation conditions. The advantage of the relative method is that it is not necessary to have accurate data regarding the emission rate and energy distribution of the irradiating source. Stability of emission characteristics is essential but this can readily be checked by repeated reference to a standard gas.

Air, nitrogen, and argon have all received a great deal of attention and their absolute energy per ion pair values for nominally 5 MeV alpha particles are reasonably well established (I.C.R.U., 1962). A summary of mean values accepted by I.C.R.U. is shown in table 8.1.

TABLE 8.1 MEAN VALUES FOR ENERGY PER ION PAIR IN AIR, NITROGEN, AND ARGON FOR 5 MeV ALPHA PARTICLES

GAS	ENERGY PER ION PAIR (eV)
air	34.98
nitrogen	36.39
argon	26.3

8.2 Ionizing sources

Early experiments using americium foil or vacuum sublimed sources mounted in a holder which produced a degree of self collimation were soon abandoned, in favour of a plane vacuum sublimed source bolted to a stainless steel backing plate and mounted in the ionization chamber such that 2π emission geometry was achieved. The arrangement is shown diagrammatically in figure 8.1.

Figure 8.1 Source mounting arrangement

8.3 Procedure

Before an experimental run, the ionization chamber and gas system were evacuated to a pressure of 10^{-6} torr and baked at 200°C for at least 48 hours. A pressure rise test was carried out on the evacuated system to check leak tight integrity and outgassing rate. Since measurements were made at gas pressures in excess of atmospheric pressure, inleaks were virtually eliminated. The experimental gas was flushed through the oil-free copper delivery tube for some minutes and was then admitted to the system. Pressure was measured by the calibrated strain gauge transducer.

The ionization current was determined at various values of collecting field, and saturation currents were determined by the usual extrapolation technique.

In the case of helium, the gas purification plant was set in operation immediately following gas entry into the chamber. A record was made of the fall in ionization current until the minimum value was reached, at which time the measurements referring to purified helium were made.

8.4 Experimental results

8.4.1 General introduction

In addition to determining values for the energy per ion pair in helium, the experiments were designed to test the effectiveness and stability of the gas purification plant under various conditions. A group of experiments

normally included measurements made with two standard gases, so that comparisons of ion yield between the standards could be used as a check that the system was operating correctly.

Ion yield data was collected in terms of the time required to collect a pre-determined charge on the integrating capacitor, and, since only relative measurements are relevant, these have not been translated into absolute current units. Each value of charge integration time was determined by calculating the mean of many such measurements made at a single value of collecting field. Extrapolation curves were drawn and the saturation ion current is presented as the 'extrapolated charge integration time' in the following results.

Virtually complete ion collection was normally achieved in all gases except air at relatively low values of collecting field. A typical extrapolation curve illustrating this point is shown in figure 8.2.

Figure 8.2 Typical current extrapolation curves

8.4.2 Comparisons of ion yield in helium and standard gases

8.4.2.1 Cylinder (unpurified) helium and nitrogen

A number of different gas cylinders containing helium were used in the course of this work, and comparisons with nitrogen indicated that the value of energy per ion pair for helium from the supply cylinders varied between about 30 and 40 eV per ion pair.

8.4.2.2 Helium, nitrogen, and air

A series of experimental runs in the three gases resulted in extrapolated charge integration times being calculated as shown in table 8.2.

TABLE 8.2 EXTRAPOLATED CHARGE INT. TIME FOR HELIUM, NITROGEN, AND AIR

GAS	EXT. CHARGE INT. TIME (sec)
helium	257.9
nitrogen	216.0
air	207.2

It may be inferred that, for the ^{non-}electronegative gases, ion collection was virtually complete since the integration time measured was constant to within $\pm 0.2\%$ for an increase of twice the collecting potential. In the case of air, measurements were made up to potentials beyond which the possibility of electrical breakdown within the apparatus became increasingly important.

Energy per ion pair values for helium were calculated by reference to each standard gas and the results are shown in table 8.3.

TABLE 8.3 ENERGY PER ION PAIR VALUES FOR PURIFIED HELIUM RELATIVE TO NITROGEN AND AIR

GAS	ENERGY PER ION PAIR (eV)	
	standard	helium
nitrogen	36.39	43.45
air	34.98	43.54

As a check on these results, the relative ionization produced in nitrogen and air was compared with the ratio obtained from their accepted values for energy per ion pair, as shown in table 8.4. Agreement to within $\pm 0.1\%$ is taken as an indication of the satisfactory behaviour of the system.

TABLE 8.4 COMPARISON OF OBSERVED ION YIELD WITH ACCEPTED VALUES FOR ENERGY PER ION PAIR FOR NITROGEN AND AIR

	accepted values	observed values
energy per ion pair for nitrogen	1.040	1.042
energy per ion pair for air		

8.4.2.3 Helium, nitrogen, and argon

A series of experimental runs in the three gases resulted in extrapolated charge integration times being calculated as shown in table 8.5.

TABLE 8.5 EXTRAPOLATED CHARGE INT. TIME FOR HELIUM, NITROGEN, AND ARGON

GAS	EXT. CHARGE INT. TIME (sec)
helium	258.4
nitrogen	214.6
argon	157.0

Energy per ion pair values for helium were calculated by reference to each standard gas and the results are shown in table 8.6.

TABLE 8.6 ENERGY PER ION PAIR VALUES FOR PURIFIED HELIUM RELATIVE TO NITROGEN AND ARGON

GAS	ENERGY PER ION PAIR (eV)	
	standard	helium
nitrogen	36.39	43.81
argon	26.3	43.26

The relative ionization produced in nitrogen and argon was compared with the ratio obtained from their accepted values for energy per ion pair, as shown in table 8.7.

TABLE 8.7 COMPARISON OF OBSERVED ION YIELD WITH ACCEPTED VALUES FOR ENERGY PER ION PAIR FOR NITROGEN AND ARGON

	accepted values	observed values
energy per ion pair for nitrogen	1.388	1.366
energy per ion pair for argon		

The following comments may be made in respect of these results and those of the preceding section:

- (i) The energy per ion pair values for helium agree less well for each standard gas than in the previous section although all results are within $\pm 1\%$ of a mean value of 43.4 eV per ion pair.
- (ii) The comparison of ion yield in nitrogen and argon do not agree with accepted values as closely as in the previous section.
- (iii) Examination of the extrapolation curve for nitrogen in the second group of experiments indicated a less satisfactory saturation collection of ions which must presumably be due to the presence of an electronegative impurity.

8.4.3 Effectiveness and stability of the gas purification plant

During the course of the experimental work, the operation of the purification plant was questioned in terms of the optimum arrangement of the traps and their degree of dependence on temperature. Additionally, with the emergence of entirely new purification trains for helium, it was necessary to determine whether an improvement was obtained when compared with the more traditional trapping reagents.

Early work had shown the importance of thorough evacuation and baking out of the system. Failure to carry out this operation properly resulted in obviously poor purification as measured by the ion yield. At least three hours continuous purification was normally required to achieve a

steady minimum ionization current in helium.

Two experiments with different purification plant systems were carried out. The first, using charcoal and calcium traps, was carried out with the flow scheme reversed so that gas emerging from the hot trap was passed through the cold trap before entering the ionization chamber. The second experiment reverted to the original flow scheme as shown in figure 7.1, but molecular sieve and hopcalite replaced the original reagents. The results are shown in table 8.8, which indicates the extrapolated charge integration time for both systems.

TABLE 8.8 EXTRAPOLATED CHARGE INT. TIME FOR PURIFIED HELIUM

PURIFICATION PLANT STATUS	EXT. CHARGE INT. TIME (sec)
gas flow 'reversed'	261.2
'normal' flow - new reagents	259.7

The apparent increase in trapping efficiency in the flow 'reversed' case is somewhat disconcerting in view of the implications for purification plants that have been used by other workers, but, in any case, represents a discrepancy of less than 1% in energy per ion pair measurements.

In an examination of the purification plant stability, measurements were made over a period exceeding 30 hours, during which time there was no significant change in the observed ion yield. No variation was observed when the molecular sieve trap refrigerant was changed from liquid air (-187°C) to liquid nitrogen (-196°C), a not unexpected result, but, more surprisingly, the hopcalite trap seemed of little value, since no variation was observed when the operating temperature was reduced to room temperature from 400°C . Of particular significance was the result obtained following isolation of the ionization chamber from the purification plant, when the expected rapid rise in ion yield as observed by other workers was not obtained. Presumably this is a function of the system cleanliness. The point is discussed in greater detail in chapter 10.

Following the testing of the purification plant, a comparative measurement of energy per ion pair for helium relative to argon produced

a value of 43.4 eV per ion pair.

8.4.4 Summary

The data presented in this chapter leads to the conclusion that W for pure helium is 43.5 ± 0.5 eV per ion pair. The stated error in the measurement includes that arising from a relative determination against a standard gas which was observed to exhibit a poor current saturation characteristic. If this reading is eliminated the error is reduced to that of the stability of the current measuring system described in the previous chapter. The use of two standard gases in determining W for helium provided a useful check that the system was operating correctly.

The efficiency and stability of the gas purification plant have been examined and found satisfactory, with the observation that direction of gas flow may be significant in ultimate attainable purity.

CHAPTER 9. ABSOLUTE MEASUREMENT OF THE ENERGY PER ION PAIR IN HELIUM

9.1 Introduction

In view of the difficulties inherent in producing an accurate measurement of the alpha particle energy emitted into the gas, the absolute determination of the energy per ion pair in helium was regarded as less important than relative measurements using gases with well established values of energy per ion pair. However, by making some assumptions, including some regarding the emission characteristics of the type of alpha particle source used, it was possible to calculate an absolute value of the energy per ion pair for ^{241}Am alpha particles in pure helium.

9.2 Source calibration

It has been shown by Yeates (1970) that the quantitative description of the alpha particle emission from a 2π planar source is complex due to the backscattered distribution from the source mount, and subsequently the accurate measurement of total energy emission rate is difficult to achieve. Yeates has carried out this study for a number of sources including a vacuum sublimed type identical to that used in the present work. By measuring a number of energy spectra at discrete angles of emission from the plane of the source varying from normal to 2° , the total energy emission rate over 2π was computed, together with the total emission rate over 2π . It has been shown from this data that the mean alpha particle energy for a vacuum sublimed source on stainless steel is reduced from 5.48 MeV for angles of emergence near normal to the plane of the source to about 5.36 MeV over 2π . It may be assumed that all similarly prepared ^{241}Am sources having a similar activity would exhibit the same reduction in mean alpha energy.

The measurement of the source emission rate used in ionization experiments was carried out using both the 2π proportional counter already described, and the ionization chamber fitted with a Frisch grid with a 90% argon/ 10% methane gas filling. Both results are shown in table 9.1.

TABLE 9.1 SOURCE EMISSION RATE MEASUREMENTS

Proportional counter:	$238.8 \pm 0.5\%$ alphas per second
Ionization chamber:	$238.7 \pm 0.3\%$ alphas per second

The ionization chamber measurements were carried out using an Ortec 118A F.E.T. preamplifier in conjunction with conventional shaping amplifiers and a multichannel analyser. An energy spectrum from the chamber is shown in figure 9.1 which also shows the peak due to a precision pulser. After subtraction of the electronic noise, the observed resolution of 69 keV F.W.H.M. is in reasonable agreement with the value of 94 keV calculated by Yeates from his low geometry extrapolations.

Figure 9.1 Alpha particle spectrum from ²⁴¹Am in gridded ionization chamber

In order to calculate total energy emission, the mean alpha particle energy has been assumed from Yeates' work to be 5.36 MeV, although the discrepancy between measured and extrapolated total count rates in his work (about 5%) leaves some doubt as to the accuracy of this figure. Certainly, the problem of very accurate total energy emission rate measurements from planar sources is complex and warrants further study.

9.3 Current measurement

The absolute measurement of ionization current is dependent on the satisfactory integration of charge on a calibrated capacitor during a known time interval, and the measurement of the potential appearing across the capacitor at the commencement and conclusion of that time interval. In the admittedly approximate system used here, the integrating capacitor was a first grade polystyrene type and was measured using a standard capacitance bridge to an accuracy of $\pm 0.1\%$. The first of the potential limits across the capacitor was taken to be zero, that is when the capacitor was short-circuited by the discharge button of the electrometer, while the second limit was determined by the system already described in section 7.4, with the electrometer output calibrated against a standard cell. The time interval was measured by the gated pulse generator as shown in figure 7.4.

In a later version of the measuring system, the polystyrene capacitor has been replaced by a standard air capacitor, the possibility of zero limit error due to charge production by switch contacts has been eliminated, and timing operations are carried out using a crystal clock which triggers a calibrated digital voltmeter to provide much greater accuracy in potential measurements.

The ionization current in pure helium was determined from the following data:

integrating capacity	= 1333 pF $\pm 0.2\%$
potential rise	= 0.910 volts $\pm 0.1\%$
integration time	= 264.8 seconds $\pm 0.2\%$

A current of $4.56 \cdot 10^{-12}$ amps was calculated from this data, indicating the production of $2.863 \cdot 10^7$ ion pairs per second in helium.

9.4 Summary of results

The energy per ion pair for pure helium was determined to be 44.7 eV per ion pair. As a check, a similar measurement was carried out using nitrogen as a standard gas, from which the energy per ion pair was determined to be 36.6 eV per ion pair.

Although these figures are corrected for background (determined to be about 0.5% of the current due to the source), they are not corrected for contributions from heavy recoils or low energy gamma radiation from ^{241}Am . These would tend to increase the apparent energy per ion pair by about 1% but, since the derived value for nitrogen is rather high, it is evident that a systematic error is present. The use of Yeates' value for the mean energy of alpha particles emitted over 2π must be held in doubt. It appears that, in view of the substantially higher number emission rate as observed with both the 2π proportional counter and the ionization chamber compared with Yeates' computer extrapolated data, the value of 5.36 MeV for the mean alpha particle energy is rather low. The spectrum obtained from the gridded ionization chamber supports this conclusion, particularly as the low energy tail is less significant.

10.1 Introduction

The necessity of using pure helium for energy per ion pair measurement has already been pointed out, and it is unnecessary to reiterate the reasons. Suffice it to say that no previous worker has paid attention to the problem of gas analysis or has made an attempt to estimate the probable purity of the helium used. In this chapter the attempts made to rectify this omission will be discussed.

10.2 The effect of outgassing of the apparatus

10.2.1 Introduction

Since most other workers examining ionization phenomena in helium have noted the rapid increase of ionization following the cessation of repurification, and since this has been attributed to outgassing of the materials from which the apparatus was constructed, the following experiments were carried out:

- (i) The increase of ionization was measured following closure of the purification system valves.
- (ii) The outgassing rate of the ionization chamber was estimated by a pressure rise measurement following evacuation of the chamber. This measurement was compared with theoretical calculations for outgassing rates of the materials concerned.
- (iii) The outgassing rate was compared with the recirculation rate employed in the pumping.
- (iv) The probable rate of increase of impurities, following closure of the purification system, was compared with the results of experiments in which known quantities of impurity gases were added to purified helium.

Each of these experiments will now be considered in turn.

10.2.2 The increase of ionization due to outgassing of the apparatus

Experiments were carried out in which helium was purified for about

12 hours after which the ionization chamber was isolated from the purification plant by closing the valves and stopping the recirculation pump. Typical results are shown in table 10.1 and figure 10.1.

TABLE 10.1 IONIZATION RELATIVE TO THAT OBSERVED IN PURIFIED HELIUM AS A FUNCTION OF TIME FOLLOWING CLOSURE OF THE PURIFICATION PLANT

TIME FROM ISOLATION OF CHAMBER (hours)	OBSERVED IONIZATION RELATIVE TO PURIFIED HELIUM
0	1
3.25	1.003
4.25	1.004
5.25	1.009
6.25	1.010
21.25	1.020
22.75	1.030
26.75	1.047

Figure 10.1 Effect on ionization current of the outgassing of the apparatus

10.2.3 The estimation of outgassing rate by pressure rise measurements

A series of experiments was carried out in which the ionization chamber was isolated from the vacuum system while evacuated. The rise in pressure was recorded by means of Penning gauges. Examination of pressure rise data of this nature can yield information regarding inleaks and virtual leaks, that is, true leaks from the surrounding atmosphere and outgassing of the apparatus. The former will be constant over a large pressure range while the outgassing rate decreases as the equilibrium pressure is approached. These measurements were considered to be a check on the normal procedure of mass spectrometer leak testing, and were carried out when the apparatus had been demonstrated to be leak tight to within the limits of the mass spectrometer used. Details of these experiments are to be found in chapter 6 and comparisons of experimental and theoretical outgassing figures are shown later in this chapter in section 10.2.5.

10.2.4 ~~Theoretical~~ Calculation of outgassing rate

Although it is difficult to apply outgassing figures to the complex apparatus used, a figure has been estimated which is unlikely to have been exceeded. By comparing the results in which the increase of ionization was observed following isolation of the purification system with gas mixture studies, an approximate cross-check on the observations may be made.

Outgassing data for some materials used in the system are shown in table 10.2, which also shows the approximate surface area and total outgassing rate for each.

TABLE 10.2 ~~THEORETICAL~~ OUTGASSING RATES (Dayton, 1961)

MATERIAL	OUTGASSING RATE (litre torr cm ⁻² sec ⁻¹)	TOTAL SURFACE AREA (cm ²)	TOTAL OUTGASSING RATE (litre torr sec ⁻¹)
stainless steel (unbaked)	10 ⁻⁷	15 10 ³	1.5 10 ⁻³
stainless steel	5 10 ⁻¹⁵	15 10 ³	7.5 10 ⁻¹¹
viton	2 10 ⁻⁸	negligible	-
P.T.F.E.	2.5 10 ⁻⁸	10	2.5 10 ⁻⁷

The most significant material in terms of surface area is stainless steel. The wide difference in the values for baked and unbaked material emphasizes the need for caution.

10.2.5 Comparison of outgassing and gas circulation rates

The steady state achieved in the dynamic equilibrium between removal of impurities and their production by outgassing is a function of the recirculation rate around the system. Since the removal efficiency of the traps is not known, it is not possible to calculate the probable level of impurities within the chamber during an experiment. The results and calculations have not been directed at specific impurities but rather have examined the gross aspects of the problem.

The gas recirculation rate was measured using a calibrated flowmeter, and was found to be not less than 1 litre per minute. Table 10.3 shows the theoretical and experimental outgassing rates together with the estimated time taken to reach an impurity level of 1 part in 10^6 in the ionization chamber without purification or recirculation.

TABLE 10.3 THEORETICAL AND EXPERIMENTAL OUTGASSING RATES AND THE TIME REQUIRED TO REACH AN IMPURITY CONCENTRATION OF 1 IN 10^6

	TOTAL OUTGASSING RATE (litre torr sec ⁻¹)	TIME TO REACH 1 IN 10^6 (min)
theoretical (unbaked)	$1.5 \cdot 10^{-3}$	0.5
theoretical (baked)	$7.5 \cdot 10^{-11}$	1.2 ^{10⁵} 2.5 ^{10⁵}
experimental	$5 \cdot 10^{-6}$	160

It is probable that, although the apparatus was baked to 200°C, the surface imperfections and only fair degree of surface polish were contributing factors which produced an outgassing figure nearer that for unbaked steel.

In the experimental arrangement, with a recirculation rate of 1 litre per minute, the gas in the 50 litre system could be totally recirculated about every 50 minutes. Hence it would be expected that the impurity

concentration would be less than 1 in 10^6 during an experimental run. Further proof of this conclusion is provided by an experiment in which the recirculation pump was stopped after purification had been carried out for some hours. The results have already been described in section 10.2.2.

10.2.6 Comparison of outgassing and impurity mixture experiments

A direct comparison may be made between experiments in the gas mixtures which arise from outgassing of the apparatus and deliberately produced mixtures of gases, although general assumptions must be made regarding the likely constituents of outgassing. Since gas mixtures are to be considered in greater detail in the following chapter, only the relevant results will be discussed here.

Table 10.4 presents results showing the ionization produced in a helium mixture relative to that in the pure gas. The admixture concentration is shown for argon, nitrogen, and outgassed impurities when the apparatus was allowed to contaminate the purified helium. The probable concentration of the total impurities in the latter case was calculated from the outgassing rates discussed in the previous sections.

TABLE 10.4 IONIZATION RELATIVE TO PURIFIED HELIUM FOR GAS MIXTURES

OBSERVED IONIZATION RELATIVE TO PURIFIED HELIUM	% OF ADMIXTURE (by vol.)		
	ARGON	NITROGEN	ESTIMATE FROM OUTGASSING
1.01	0.06	0.015	0.003
1.015	0.092	0.02	0.004
1.02	0.1	0.03	0.006

The conclusion to be drawn from this comparison is that the estimated outgassing rate is rather low, although certainly less than an order of magnitude low. It should be emphasized that such experiments are probably only accurate to within an order of magnitude as far as outgassing is concerned, and in any case only refer to the total increase in impurity concentration as measured by a Penning gauge. Since no means for analysing the constituents of outgassing was available, no account was taken of the relative concentrations

of different impurities, although it is expected that nitrogen and possibly water vapour will be the most important. In any case, the relative composition of the outgassing mixture is probably of little significance in view of the comparative insensitivity of the Jesse effect to the likely contaminants.

10.3 Gas purity measurements

10.3.1 Introduction and general principles

It is evident from preceding discussion that the measurement of gas purity is a necessary part of ionization studies in helium. Formerly it was thought sufficient to accept the criterion of minimum ionization as being an indication of pure gas, but, from the spread of experimental values for the energy per ion pair in helium, this is evidently not enough. It is certainly true that, for a closed circulation system, a condition of dynamic equilibrium will exist within the ionization chamber, as a result of the function of the purification plant and the desorption of impurities from the apparatus. Whether the resulting impurity concentration is sufficient to influence the production of ionization has been an open question, and no attempt has previously been made to measure this.

Any technique capable of identifying and measuring the components of a mixture requires both means of separation of the components and a detector that is sensitive to them. Since permanent gases with small mass numbers are the most frequently encountered impurities in helium, and certainly the most abundant, it was considered possible that mass spectrometry could well provide the necessary analytical tool. This method with relatively modest equipment is capable of unit separation of masses to about 45, and is therefore ideally suited for examining permanent gas concentrations. The sensitivity of typical small instruments, however, is such that approximately 10 parts per million is the lowest detectable concentration. Large instruments with much greater sensitivity are available but were impracticable for the purpose of this work as will be discussed later.

A technique which relies upon separation of gas components with respect to time is that of gas phase chromatography, in which a quantity of the unknown gas mixture is admitted to the top of a separation column and carried

down the length of the column by a carrier gas. Separation is achieved depending on the characteristics of the column, which may be molecular sieve or any suitable adsorbent. The detector is normally exposed to the carrier gas flow, and, at intervals, to the components of the mixture. Although a number of detectors employing varying physical principles have been used for gas chromatography, most of them are suitable only for the detection of organic molecules. The most satisfactory detector for use with permanent gases using helium as the carrier gas has proved to be a modification of the Lovelock (1958) ionization detector, which will be discussed in further detail in section 10.3.3.

10.3.2 Mass spectrometry

The limiting characteristics of mass spectrometers include their resolving power and their minimum and maximum operating pressures in the case of gas analysis. With permanent gases being the most serious contaminants in helium, the resolving power of nearly all instruments was considered to be adequate for a quantitative analysis to be carried out, but the minimum detectable partial pressure and the maximum operating pressure, together with the vacuum systems used, virtually precluded a satisfactory analysis of helium to the desired impurity concentration of a few parts per million. Two instruments will be described.

The existence at the University of an extremely sensitive spectrometer (A.E.I. M.S.12C) of high resolving power, prompted an investigation into the possibility of carrying out batch analyses by sampling into a suitably prepared vessel and transfer to the spectrometer. It was considered that this operation, although difficult, could result in a pure sample being maintained without additional contamination for the period during which a transfer was carried out. The spectrometer itself, however, was used almost exclusively for high molecular weight chemical analysis where resolving power, rather than sensitivity, was of the greatest importance. In view of this, the partial pressures of permanent gases within the instrument were relatively high and subject to considerable fluctuation, a combination of factors which precluded satisfactory analyses.

The second instrument was an A.E.I. M.S.10 spectrometer which allowed continuous gas sampling from the purification plant. Unit mass resolution

was available up to about mass number 85 and the minimum detectable partial pressure was 10^{-10} torr. Later versions of the instrument are an order of magnitude better in sensitivity. With a maximum operating pressure of 10^{-4} torr, the lower limit of impurity concentration measurable was about 1 part per million. Since the gas to be sampled was at a pressure exceeding that of atmospheric, a sample introduction system was used in which a small quantity of gas was allowed to leak through a sintered disc into the analysing head, the remainder being bypassed to a rotary pump. The main vacuum pumping system was common to the ionization chamber and the mass spectrometer and the relatively poor vacuum attained with this equipment (about 10^{-6} torr) limited the measurable lower impurity concentration to about 1%. In order to reach the maximum sensitivity of the spectrometer, it is necessary to use molecular sieve sorption pumps and getter ion pumps to achieve a suitable order of vacuum. The use of getter ion pumps is almost precluded in a continuous sampling mode because of their limited life and high cost.

The immediate conclusion to be drawn is that the instrument described is most suitable for analysing gas mixtures where the concentration of the constituents is not less than 1% of the total. A further difficulty is the necessity of carrying out an accurate total pressure measurement with, for example, an ionization gauge. Such an instrument was not originally available but has since been installed, and carries out the additional task of acting as a safety cut-out to prevent a burn-out of the spectrometer filament in the event of an excessive pressure being applied. With the short-comings of the basic equipment, on loan to the author for a short period only, few conclusions could be reached regarding the absolute purity of the helium in use, other than the general conclusion that it was probably not grossly contaminated to more than a few percent of impurities.

10.3.3 Gas chromatography

Although the gas chromatographic technique proved to be unsatisfactory, the principles and philosophy of the system used have been included in this section together with some comments regarding the possibility of improving the response of the detector.

The principle of radioactive ionization techniques to detect the elution of sample constituents from the column of a gas chromatograph was first suggested by Lovelock (1958,1961) and has been elaborated by many workers. The most usual application is concerned with the detection of organic molecules in a carrier stream of argon. A radioactive isotope (usually ^{90}Sr) produces metastable excited argon atoms which are capable of ionizing by collision the organic molecules in the stream. The observed ionization current varies with the concentration and type of constituent in the carrier stream, separation being achieved by a suitable chromatographic column. The problem of permanent gas analysis is treated by using helium as the carrier gas since it has a metastable excited state of a higher energy than the ionization potentials of most common gases. Neon is a special case since its ionization potential exceeds that of the metastable helium state, but its presence in a helium carrier may still be detected by the reduced ionization at the detector resulting from direct competition with helium.

The usual form of ionization detector employs a few millicuries of ^{90}Sr surrounding a rather empirically designed electrode system. Two types of detector are commonly used, both called argon ionization detectors after the usual carrier gas. The macro-argon detector is used with conventionally packed chromatography columns, while the micro detector is one in which a purge flow of carrier gas is used to contain elutants within the high field strength region. The characteristics of the latter have been investigated quantitatively by Collinson et al. (1965). Later work by Hartman et al. (1966) described a detector particularly for use with helium as the carrier gas. About 250 mCi of tritium is used in the form of a circular foil, which is also the collecting electrode of the parallel plate system enclosing a sensitive volume of 160 μl . Sensitivities of about 1 part in 10^9 have been claimed for the detector. In the present work a conventional macro-argon detector with 10 mCi of ^{90}Sr was used, mounted within a vacuum-tight brass chamber to eliminate problems of contamination from leaks.

The complete analysis system incorporating facilities for detector calibration is shown in figure 10.2. Calibration of the detector can be carried out by introducing known concentrations of helium admixtures, using the flowmeter dilution train as shown in figure 10.3 which permits

dilutions down to 1 part in 10^8 to be obtained.

Figure 10.2 Chromatographic analysis system

Figure 10.3 Flowmeter dilution train

Purified helium is allowed to flow up the molecular sieve chromatographic column until a sample is introduced by the sampling valve. The type of valve used incorporates a purge flow which effectively eliminates the possibility of accidental contamination during sample introduction. As a check that the usual purification train is operating efficiently, provision is made for a liquid helium trap which is the most efficient means of trapping all impurities in helium.

The construction of the complete system was of glass and stainless steel with P.T.F.E. used for valve seals. The flowmeters were calibrated using a soap film flowmeter.

Following outgassing of the pipework it is necessary to maintain a continual flow of purified helium to prevent adsorption of contaminants, an operation estimated to cost at least £30 per week for helium alone.

The procedure for commissioning the system was initially that of determining the characteristics of the detector under various conditions. Fixed values of high voltage were available from the detector control unit which also incorporated a d.c. amplifier with switched attenuators and a variable back-off facility. The dependence of detector current on helium flow rate at various values of applied voltage was determined as shown in figure 10.4, indicating a limited dependence at flow rates exceeding about 400 ml minute⁻¹. The relatively strong dependence of current on detector voltage is also shown in the figure, a value of 750 volts being chosen as the detector voltage for all measurements, to prevent the occasionally observed high currents due to flashover in the detector.

Calibration of the detector for various impurity concentrations proved to be unreliable as may be seen from figure 10.5, which presents two sets of results each for argon and nitrogen concentrations below 50 parts per million. The only significant difference between conditions for curves (a) and (b) was that flow rates for the former were generally greater than those for the latter, although both were within the region of independence of detector current on flow rate. The lack of reproducibility was traced to diffusion of impurity gas into the purified helium flow. In particular, an order of magnitude decrease in detector current was observed when nitrogen was admitted to the impurity mixing train, although no nitrogen should have

Figure 10.4 Detector current as a function of helium flow rate at various values of detector voltage

Figure 10.5 Detector calibration for impurities

reached the detector. The ~~necessity~~^{necessity} of a fresh approach to this problem is quite evident and is reinforced by the unsatisfactory results obtained using the chromatographic column. The column was fabricated from molecular sieve type 5A ground and sieved to select 50-60 mesh size particles, which were packed into a stainless steel column of about 40 cm length and 3 mm bore. A heating tape allowed regeneration of the adsorbent when saturated. No results are presented for the column since calibration proved impossible but an elution time of about 30 seconds was observed for nitrogen.

10.3.4 Water vapour

The most sensitive and satisfactory method of measuring water vapour concentration in a gas stream is that of the electrolytic hygrometer. Proposed by Kiedel (1959) and improved by Goldsmith (1967), the cell consists of two fine platinum wires wound alternately at close spacing on a grooved P.T.F.E. rod coated with phosphorus pentoxide, the whole unit being housed in a glass cell. As the gas passes through the cell, water vapour is absorbed and electrolysed to hydrogen and oxygen by a potential maintained across the platinum wires. By Faraday's Law of Electrolysis, 0.5 g mole of H_2O requires 96,500 coulombs. For a gas flow of 100 ml minute⁻¹ at room temperature and atmospheric pressure, the current through the cell will be 13.2 microamps per part per million of H_2O , assuming 100% water vapour absorption efficiency.

The instrument supplied by G.E.C. Limited was coupled to the output of the purification plant using stainless steel tubing with a glass flowmeter calibrated for helium by means of a soap film flowmeter. A commercial needle valve was found to be satisfactory for fine flow control. The output of the flowmeter was passed through a low vapour pressure silicon oil to prevent back diffusion of water vapour.

During open line testing of the purification system it was observed that, after running the plant for some hours, the water vapour concentration fell to about 0.5 parts per million and remained constant for a number of days. Since the sampling point of the hygrometer was chosen to be the output of the molecular sieve trap, that is, directly before the purified gas entered the ionization chamber, measurements were also made with the

molecular sieve at room temperature, when concentrations of about 10 parts per million were observed. This is an indication that the sieve is only lightly loaded and extrapolation to liquid air temperature produces a probable water vapour concentration of considerably less than 0.5 parts per million.

11.1 Introduction

The purpose of this series of experiments was primarily the corollary of gas analysis, that is, the deliberate contamination of purified gas. It was also suspected that the purity of the helium achieved was considerably better than that used in Jesse's work and therefore that a re-examination of his admixture results was necessary.

11.2 Procedure

A sampling bomb of known volume was filled with the additive gas to a pressure measured by the strain gauge pressure transducer. Opening of an isolation valve permitted the additive to mix with purified helium within the ionization chamber. The ratio of volumes of sampling bomb and ionization chamber allowed the impurity concentration to be determined.

At the time of these experiments, an analytical mass spectrometer was not available to check the partial pressures of the mixture. It is recommended that this be carried out in future work to avoid the errors which could arise as a result of selective adsorption on the walls of the apparatus.

11.3 Results

The results are summarised in table 11.1 and shown, together with Jesse's results in figure 11.1.

11.4 Comparison with Jesse's results

The main feature shown by the comparison is that the assumption that the present purified helium is purer than Jesse's is apparently correct. This is borne out chiefly by the additional saturation increase in ionization

TABLE 11.1 IONIZATION RELATIVE TO PURE HELIUM FOR LOW CONCENTRATIONS
OF IMPURITIES

% IMPURITY	IONIZATION RELATIVE TO PURE HELIUM	
	NITROGEN	ARGON
0.05	1.034	1.008
0.1	1.074	1.020
0.15	1.188	1.05
0.2	1.290	1.17
0.25	1.357	1.299
0.30	1.393	1.370
0.35	1.425	1.41
0.40	1.429	1.435
0.45	-	1.444
0.50	-	1.47
0.55	-	1.474
0.60	-	1.476

Figure 11.1 Ionization relative to pure helium for low concentrations
of impurities

observed in the present experiments. Further, the displacement along the impurity concentration may be taken as additional proof. The puzzling aspect of this work is the apparent tail near the origin. It may be argued that Jesse's experiment was insensitive to this region so that this is a true effect. That it may be an artefact due to selective adsorption of small admixtures of impurity on the containing walls can only be disproved by direct analysis of the mixture, but, in any case, it is most unlikely. A possible explanation lies in the distinction between Penning ionization by the metastable and non-metastable states. At low concentrations of impurity, the metastable states will be sufficiently long lived to allow collisions, while at higher concentrations (above about 1 in 10^3), it is probable that non-metastable excited atoms will encounter an impurity during their lifetime of about 10^{-7} seconds. Following a collision, ionization of the impurity atom takes place with a probability approaching unity. Hence the saturation increase in ionization refers not only to metastable states, but to some non-metastable states as well. This result has been predicted theoretically by Erskine (1954) who found that the total observed increase in ionization could only be explained if all excited states led eventually to ion pairs.

At impurity concentrations below that which would result in a collision between an impurity atom and a non-metastable excited helium atom during the lifetime of the latter, there would, presumably, be a less marked effect since only the metastable state would have a sufficiently long lifetime.

An additional effect which results in ion production from excited states is that of dimeric ion formation resulting mainly from $n > 3$ states. This will be marked at higher pressures where these states do not decay before collision with a neutral atom.

11.5 Penning ionization cross-sections

From gas mixture curves of the type shown in figure 11.1, it is possible to derive comparative values for Penning ionization cross-sections. Jesse, referring to metastable atoms assumed that, in pure helium, metastable states are destroyed by radiative processes, while in contaminated helium

at the saturation value all metastable states led to ion pairs. At half the saturation value the two destructive processes occur in equal numbers, and the probability of a collision producing an ion pair is equal to the probability of the metastable state losing energy without producing an ion pair. This may be represented by:

$$\sigma_i N_i v_i = \sigma_d N_{\text{He}} v_d$$

where σ_i and σ_d are the cross-sections for the two possible reactions, N_{He} and N_i are the numbers of atoms per ml of helium and of the impurity gas respectively, and v_i and v_d are the relative velocities of approach of the particles in the two cases.

The results of Jesse's analysis are given in table 11.2 together with results derived from the present work on the same basis.

TABLE 11.2 RELATIVE CROSS-SECTIONS FOR DESTRUCTION OF HELIUM METASTABLES

AUTHOR	ARGON IN HELIUM (σ_i/σ_d)	NITROGEN IN HELIUM (σ_i/σ_d)
Jesse	$9.3 \cdot 10^3$	$8.6 \cdot 10^3$
Harris	$5.6 \cdot 10^2$	$8.12 \cdot 10^2$

The discrepancy is made more serious by reason of Jesse's use of a rather low value for σ_i due to Biondi (1953). It may be that Jesse is quite right in assuming that metastable states are responsible for the increase in ionization that he has observed, indeed his results agree quite well with some other work specifically designed to measure metastable de-excitation cross-sections, but for the triplet state rather than the singlet (Benton, 1962). These optical experiments are compared in table 11.3 with results from crossed beam techniques (Sholette, 1962), together with Jesse's results and those from the present work. The total ionization experiments are both normalised to Benton's value for the triplet cross-section.

In the experiment performed by Jesse and Sadauskis, it was postulated that singlet states were mainly responsible for the observations. If this is the case, then comparisons with cross-sections measured by other techniques must be treated with extreme caution in view of the wide discrepancies between crossed beam and optical techniques. The lack of

specificity of total ionization experiments makes this interpretation extremely difficult.

TABLE 11.3 PENNING IONIZATION CROSS-SECTIONS ($^{\circ}\text{A}$)²

	$^{\circ}\text{Ti}$				$^{\circ}\text{Si}$			
	Benton	Sholette	Jesse	Harris	Benton	Sholette	Jesse	Harris
argon	6.6	8	6.6	6.6	55	8	6.6	6.6
nitrogen	6.4	7	6.1	8.6	-	7	6.1	8.6

CHAPTER 12. CONCLUSIONS AND SUMMARY OF WORK IN PROGRESS

12.1 Introduction

The purpose of this chapter is to summarise the work carried out as a result of this research programme with emphasis placed on the particular areas which require further attention. Some suggestions are made for future treatment.

12.2 Source calibration

The unexpected difficulties arising from source calibration studies tended to reduce the significance attached to absolute measurements of energy per ion pair in this work. Although the phenomenon of backscattering of alpha particles has long been known, detailed studies of this effect for specific source backings have not previously been carried out. In the work already published (appendix 2 to this thesis), a technique using a silicon surface barrier detector was used to investigate the angular distribution of alpha particles emitted from a planar source. The uncertainty in total energy emission rate is indicated by the 2% difference between computer extrapolated and 2π counter measurements. The resulting decrease of the mean energy of emitted alpha particles, however, is certainly not less than about 2% and may be considerably more. The particular problem of the energy distribution of particles emitted at glancing angles from the plane of the source is one which is specific to the source backing and surface topography. The subject deserves some further attention, possibly from the applied aspect in which low angle scatter may be used to gauge the degree of surface smoothness and topography.

There is little doubt that absolute number emission rate measurements over 2π are best carried out using a gas flow proportional counter. This well established technique has been reconsidered to include high counting rate measurements (appendix 1 to this thesis).

It is difficult to reconcile these conclusions, which in any case only apply to sources with the minimum self absorption, with the apparent ease

with which Bortner (1954) carried out total energy emission rate measurements using a 2π proportional counter only.

With the intention of carrying out gas ionization experiments at reduced pressure, reduced energy alpha sources were produced by evaporating a gold absorber directly on the active area. The various types of reduced energy sources have already been reviewed, but the difficulties of production and calibration were relatively few, provided that a considerable reduction in energy was not required. However, in order to meet the design parameters of the experiment within the limitations of the ionization chamber, an extremely high (>5 MeV) energy loss in the absorber was required. Measurement of ion energy in this region is not easy, and, in particular, the use of silicon surface barrier detectors is precluded because of their gold window and oxide layer thickness. Additionally, there is uncertainty regarding the value of the energy per electron-hole pair in the detector for the energies and types of ions involved. It is clear that a high proportion of ions will be singly charged or neutral. The method of energy measurement in this range is open to question. Possibly a time of flight technique may prove valuable.

Although energy calibration experiments were admittedly difficult, some measurements using reduced energy sources were made using both the alpha spectrometer and the ionization chamber. The spectrometer measurements showed that energy straggling soon became important, although there was apparently little evidence of high energy alpha particles penetrating through the gold absorber. Later work tended to show that some high energy breakthrough was evident (Muluneh, 1970). Confirmation of this effect, which could be due to source material flow or damage, needs to be sought. The ionization chamber experiments clearly showed that, over 2π , a considerable proportion of emitted alpha particles were at relatively high energies. This factor, combined with the very large observed energy straggling led to a reconsideration of the whole topic of alpha particle absorption, with particular emphasis being placed on almost complete absorption in a solid. A paper has been published (appendix 3 to this thesis) reporting some preliminary results in which energy straggling is shown to decrease as the particle approaches the region of total absorption in the solid film. An explanation of this effect, not previously reported in solids, has been given in terms of the variation of stopping power with

energy at low energies. Additional studies with fission fragments from a spontaneous fission source (^{252}Cf) have confirmed these conclusions, although studies of fission fragments using solid state detectors are complicated by pulse height defects arising from interactions within the detector.

Many of the source calibration difficulties would be overcome by the use of a suitable monoenergetic source such as is available from an accelerator. Work to allow a Van de Graaff accelerator to be used in conjunction with the ionization chamber is presently nearing completion. The low energy ions available from this facility will enable reduced pressure experiments to be carried out with greater ease.

12.3 Measurement of ion yield

The measurement of ion yield has been carried out using conventional current measurement techniques, although less importance has been attached to absolute values. Instead, it was decided to adopt a relative measurement and, in particular, a cheap automatic system was developed which allowed periods of operation without attention. Since this method had obvious disadvantages in the timing method and sensing of potential limits, the apparatus was recently updated to allow highly stable and accurate timing from a crystal clock, with direct potential limit display on a calibrated digital voltmeter. Pulse collection methods were only carried out in argon/methane as a result of the poor drift velocities achieved in other gases with the high pressures and chamber dimensions involved.

12.4 Helium measurements

In view of the history of energy per ion pair measurements in helium, it is hardly surprising that a note of caution enters in any claim that may be made regarding a new measurement. However, some data has been presented in the course of this work which would appear to indicate an improvement in what has known to be a difficult problem. The single factor that has been cited as responsible for lack of reproducibility in helium measurements has been that of gas purity. Although no direct measurement

of gas purity was made, other than for water vapour, it has been shown by indirect techniques that an extremely high degree of purity was achieved and adequately maintained for long periods. This result is directly in conflict with the work of Jesse (1955b) and, as expected, a higher value for the energy per ion pair was recorded in the purer gas. This conclusion regarding gas purity was strengthened by the results of gas mixture experiments, which themselves revealed an effect which has not hitherto been reported. The gas mixture studies almost certainly reflect the relative significance of metastable and non-metastable excited states in helium. Further work including variable pressure mixtures could resolve this tentative explanation.

12.5 Summary

The measurements of W for pure helium relative to standard gases can be accepted with greater confidence than the absolute values in which a systematic error is present. The relative measurements of W have given a value of 43.5 ± 0.5 eV per ion pair, while the absolute value was calculated to be 44.7 eV per ion pair. The doubt regarding source energy emission rate is almost certainly responsible for this discrepancy.

Gas mixture studies have indicated that non-metastable excited states of helium atoms are responsible in part for the Jesse effect which has previously been attributed solely to metastable states.

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Abstract The accurate measurement of high emission rates of alpha radioactive sources has been neglected up to the present time. In order to resolve this difficulty a new 2π proportional counter was developed, capable of measurement up to at least 5×10^5 alpha particles per second into 2π

geometry. Careful attention was placed on the factors limiting counting speed, notably mechanical and electrical design of the counter and preamplifier circuitry. The choice of counting gas was also critical for high speed applications. The calibration of the counter was checked using a certified standard source from the Radiochemical Centre emitting 1.029×10^5 alpha particles per minute. The measured emission rate of the source agreed to within $\pm 0.1\%$ of this figure.

1 Introduction

As part of a programme for determining absolute values of the energy per ion pair in gases using alpha particles, it was necessary to develop a fast counter operating in the proportional region. This was required for the accurate standardization of alpha particle sources which emit up to 5×10^5 particles per second into 2π geometry.

A schematic diagram of the counter is given in figure 1. The dimensions were chosen to satisfy the following requirements. Firstly, sources of 1 cm diameter and of various thicknesses were to be accommodated. Secondly, it was

necessary to provide a sensitive volume of sufficient size to ensure that, at all angles of emission, the number of primary ions produced would be sufficient to give rise to a pulse above the discriminator level. Thirdly, since the maximum count rate attainable in a counter is primarily determined by the transit time of the electrons from the primary track to the anode, this distance was reduced to the minimum value consistent with other requirements.

2 Counter design

The main body of the counter was made of copper. The sensitive volume was cubical (and not cylindrical) in shape, in order to approach 2π geometry with as uniform an electric field as possible. For the same reason, a straight wire rather than a loop anode was used. A negative polarizing potential was applied to the main body of the counter rather than a positive potential to the anode in order to reduce the noise

Figure 1 Schematic diagram of the proportional counter (gas inlet and outlet, not shown in the cross section illustrated, is effected through the top of the chamber)

voltage across the coupling capacitor between the counter and the preamplifier. The anode was made from 0.005 cm diameter stainless-steel wire and was shielded at each end by a conventional system of guard and field tubes.

An emitter follower preamplifier (figure 2) with a gain of unity was situated adjacent to the counter, but screened from it by a metal shield.

Figure 2 The emitter follower and h.t. smoothing circuits

The pulses were then amplified without pulse shaping and counted on a fast scaler. In order to be able to count at high rates, it was necessary to obtain the shortest possible pulse length with a high signal-to-noise ratio. The pulses were therefore 'quenched' by using a low value load resistor of 2.2 kΩ. The value of the coupling capacitor used was 220 pF. These values gave a slight overshoot which ensured that the pulse fell below the discriminator level of the scaler at a rapidly decreasing part of the pulse. Any increase in overshoot caused an increase in plateau slope. Using these RC values, the count rate-voltage plateau had a slope of 0.005% per volt at an operating potential of 2520 v. The count rate was found to increase by 0.013% per unit change in the gain at an amplification of 800 on the main amplifier.

3 Operation

The maximum voltage at which it is convenient to operate a proportional counter for alpha particles is determined by the voltage at which beta particles are detected. In order to ensure that no pulses due either to beta particles or gamma radiation would be counted when measuring an alpha particle source, a test was made using a strontium-90-yttrium-90 source.

Three gases were tested for use in the counter: methane (purified to greater than 99%); 90% argon-10% methane; 96% helium-4% isobutane. Typical pulse shapes obtained using these gases are shown in figure 3, and the relevant parameters of these gases and of the corresponding pulse shapes are summarized in table 1. The rise times and fall

Figure 3 Typical pulse shapes obtained: (a) pure methane; (b) 90% argon-10% methane; (c) 96% helium-4% isobutane. The horizontal scale is 10^{-7} s per division

times are measured from the position of the peak to the intersect of a straight line drawn along the leading (or trailing) edge and the base line. Pulses in both the argon-methane and in the helium-isobutane gas mixtures varied in both length and complexity. Campion (1968) also observed complex pulses using the argon-methane mixture. No satellite pulses were observed in methane using operating potentials up to 3200 v. Pure methane gas was therefore chosen as the most suitable gas for the experiments described in this paper.

The resolution time of the counter with its associated electronics was measured using the double source method (Kirby 1964). It was measured using sources with a count rate of $5 \times 10^4 \text{ s}^{-1}$ and $5 \times 10^5 \text{ s}^{-1}$, and values of 445 ± 1 and 447 ± 1 ns respectively were obtained, demonstrating the relatively slight dependence of resolving time on count rate. An error of 5 ns in the measurement of resolving time at a count rate of about $2 \times 10^5 \text{ s}^{-1}$ would result in an alteration of 0.1% in the measured count rate. The factors limiting the resolution time includes both collection times in the counter and the speed of the electronics.

The space charge or 'sheath' effect caused by the integrated effect of the positive ions moving towards the cathode did not appear to have a detrimental effect at the count rates used.

The counter system proved to be very sensitive to both transmitted electrical noise, and the noise from the mains supply. However, on placing the whole system in a screened room with radio-frequency filters, the background count rate was about 10^{-2} s^{-1} .

The errors in the count rate which may be associated with measurements made using this counter may be summarized as follows:

- (i) Background radiation: this will be negligible for sample count rates above 100 s^{-1} .
- (ii) Position on amplification plateau $\pm 0.1\%$.
- (iii) Error arising from resetting the h.t. $\pm 0.1\%$.
- (iv) The correction for resolving time; the error associated with this correction will be less than $\pm 0.1\%$ at 2×10^5 counts per second.

The calibration of the counter was checked using the strongest available standard source of americium-241. This

Table 1 Comparison of the counting gases

Gas	V	E.H.T. (v)	Rise times (ns)	Fall times (ns)	Total pulse length (ns)
Purified methane 99%	10†	2520	150	230	380
90% argon-10% methane	4†	1800	170 → 300	300	470 → 600
96% helium-4% isobutane	1‡	1320	170 → 350	240 → 650	410 → 1000

V , approximate drift velocity ($\text{cm } \mu\text{s}^{-1}$) at $E/P = 1.5 \text{ v cm}^{-1} \text{ torr}^{-1}$

† Bortner *et al.* 1957. ‡ Neilsen 1936.

Centre, Amersham, of 1.029×10^5 alpha particles emerging per minute from the front of the source. The maximum overall error was stated to be less than $\pm 3\%$. When this source was measured in the counter described above, the emission rate agreed with that of RCC to within 0.1%.

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APPENDIX 2.

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**A STUDY OF THE ANGULAR DISTRIBUTION AND
ENERGIES OF ALPHA PARTICLES EMITTED
BY ^{241}Am SOURCES**

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A STUDY OF THE ANGULAR DISTRIBUTION AND ENERGIES
OF ALPHA PARTICLES EMITTED BY ^{241}Am SOURCES*

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The angular distribution and energies of alpha particles emitted from both a vacuum sublimed and a silver-palladium coated ^{241}Am source were examined with a low geometry spectrometer

using a surface barrier detector. Computer programmes were developed to allow integration of data over 2π sterad. Data from the above two sources is presented.

1. Introduction

An alpha particle source does not emit particles isotropically either with respect to the number emitted, or with respect to their energy. The two factors which determine this anisotropy are self-absorption in the source, and back-scattering by the source mount. This latter effect, which is of particular importance at small angles of emission from the plane of the source, results in the emission of alpha particles of reduced energy. The magnitude of this effect is dependent also on the material and the smoothness of the surface of the source backing¹.

This work was part of a University of Surrey M.Sc. project submitted by D. Yeates.
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Fig. 1. Schematic diagram of the low geometry spectrometer.

The experiments described in this paper were carried out in order to determine various parameters relating to alpha particle emission by a vacuum sublimed source and by a thick alpha particle source.

2. The low geometry spectrometer

A schematic diagram of the low geometry spectrometer is shown in fig. 1. It consists of a vacuum chamber in which an alpha particle source faces an Ortec grade A silicon surface barrier detector at a distance of 13 cm from the detector. The alpha particle source can be rotated so that the angle between the plane of the source and the plane of the detector may be varied from 0° to 90° . The sensitive area of the detector and hence the solid angle geometry of the system was accurately defined by means of a collimator on the front of the detector. Two further collimators were used on the central column to ensure that no alpha particles scattered from the walls of the chamber, or from the central column, could reach the detector.

The best detector resolution measured with this detector was 15.7 keV fwhm for the 5.486 MeV alpha particle peak from ^{241}Am .

The pulses from the detector were analysed using a conventional 400 channel pulse height analysis system. Pulse spectra were obtained for a range of angles of emission between 0° and 90° , and the system was calibrated so that the energy corresponding to each channel was known accurately, (a correction was made in this calibration for the energy lost by alpha particles in the gold window of the surface barrier detector). An extrapolation procedure was used in the computer programme to correct for the finite discriminator level of the pulse analysis system.

3. The alpha particle sources

Two sources of ^{241}Am alpha particles were used in the studies described in this paper. In experiment A, the source was a thin layer of ^{241}Am which

g. 2. Diagram to illustrate the variation of the total solid angle of emission as a function of the angular emission.

was vacuum sublimed on to a stainless steel planchette. In experiment B, the source consisted of a matrix of ^{241}Am in gold on a silver base and sealed with a 2 mg/cm^2 coating of 94% silver and 6% palladium.

4. Experimental results

The observed spectra were first corrected for different counting times and for background. They were then corrected for the variation in the total solid angle of emission as a function of a given angle of emission, (fig. 2). These corrected spectra for the thin source are shown in fig. 3, and the corresponding results for the thick source in fig. 4. The change in the spectrum with angle of emission even in a thin source is immediately

Fig. 3. Corrected spectra for the thin alpha source.

Fig. 4. Corrected spectra for the thick alpha source.

apparent from the first of these diagrams. The second shows the interesting phenomenon of a significant number of low energy alpha particles being emitted over the angular range 20–65°. We have not been able to formulate a convincing mathematical theory concerning the origin of these low energy particles. Computational summation of all of the particles emitted over 2π ,

Fig. 5. Total number spectra over 2π for the thin source.

Fig. 6. Total number spectra over 2π for the thick source.

from each family of curves, resulted in figs. 5 and which relate to the total number spectrum of the alpha particles emitted over a hemisphere for the thin a thick source respectively. A further parameter obtain

Fig. 7. Angular distribution of alpha particles as seen by a sm detector — thin source.

Fig. 8. Angular distribution of alpha particles as seen by a sm detector — thick source.

Fig. 9. Angular distribution of the mean alpha particle energy — thin source.

Fig. 10. Angular distribution of the mean alpha particle energy — thick source.

in these results was the variation in the total count rate observed by a detector as the angle of emission is varied. These results are given in Figs. 7 and 8 for the thin and thick source respectively. The sharp rise in the alpha particle emission rate at low angles is clearly significant.

The total energy emission was obtained as a function of angle by multiplying the number of counts observed in each channel by the energy corresponding to that channel number. Summing over the total solid angle gave the total energy emission. The mean energy of the alpha particle as a function of the angle of emission was also obtained (Figs. 9 and 10).

Table 1 summarises the results of these two experiments. The second line of this table gives the total emission rate for each of the sources obtained from independent measurement using a 2π gas flow proportional counter. This is in excellent agreement with the results obtained using the low geometry spectrometer. It would therefore appear that the method of calculations used in the results described in this paper is valid. Lines 4, 5, and 6 give the fwhm resolution at 90° and the fwhm of the total count spectrum together with the energy of emission at 90° . The difference between the fwhm as measured at 90° and the fwhm of the total spectrum over 2π is clearly small. The final figures on this table give the total energy emission rates for each of the sources.

TABLE 1
Summary of results.

	Experiment A	Experiment B
Number of spectra between $0-90^\circ$	13	15
Total count rate using 2π proportional counter	$3.143 \times 10^6 \text{ min}^{-1}$	$1.444 \times 10^7 \text{ min}^{-1}$
Total count rate using low geometry spectrometer	$3.009 \times 10^6 \text{ min}^{-1}$	$1.438 \times 10^7 \text{ min}^{-1}$
Fwhm at 90°	59 keV	390 keV
Fwhm of total count spectrum	94 keV	720 keV
Energy of peak at 90°	5.486 keV	4.888 keV
Total energy emission rate	$1.615 \times 10^{10} \text{ keV min}^{-1}$	$5.612 \times 10^{10} \text{ keV min}^{-1}$

As an additional check on the accuracy of the computer programme, data was generated from an hypothetical experiment in which it was supposed that identical spectra were obtained at each of 14 different angles. The total number of alpha particles emitted calculated by the computer programme using these spectra should be, and was, identical to the number calculated using conventional formulae based on a single measurement with a small detector. The agreement, was, in fact, perfect, to within the accuracy of the arithmetical calculations in the computer. This adds additional confidence to the assumption that the method of computation is satisfactory.

5. Conclusions

The results described in this paper illustrate variation in the mean energy of the alpha particles emitted and the variation in the number of alpha particles emitted, both as a function of the angle of emission. The number and energy vary so much with angle of emission that large errors can be introduced in calculations of total emission rates based on single measurement. This is true both for thin and thick sources.

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ENERGY STRAGGLING OF ALPHA PARTICLES IN THICK ABSORBERS*

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Measurements have been made of the energy straggling of ^{241}Am alpha particles through thick aluminium absorbers. A region of increasing straggling has been shown to be followed by a maximum and a region of decreasing straggling towards the end of the alpha particle range. An explanation of the reduction in energy straggling or "energy-bunching" effect has been sought by reference to low energy stopping power data.

1. Introduction

As part of an investigation of emission characteristics from alpha particle sources, the energy straggling of alpha particles has been studied near the end of their range. There have been few publications on alpha particle straggling in solids¹⁾ or in gases^{2,3)}. Such measurements have become less difficult recently, with the availability of high resolution semiconductor gold surface barrier detectors and of very thin vacuum sublimed sources having little self-absorption.

2. Apparatus

The experimental chamber is shown diagrammatically in fig. 1. Two vacuum sublimed sources were used, both types available commercially from the Radio-

chemical Centre, Amersham. The first used ^{241}Am only, while the second was a calibration source using a mixture of ^{239}Pu , ^{241}Am and ^{244}Cm . A conventional diffusion pump system was used to evacuate the system to less than 10^{-5} torr. Liquid nitrogen-cooled traps in both the high vacuum and roughing vacuum lines prevented backstreaming of oil vapour from poisoning the detector, and contamination of absorbers or sources. Previous work had shown contamination to be of possible significance in high resolution studies⁴⁾. The surface barrier detector had a nominal active area of 25 mm^2 and a depletion depth of $100\text{ }\mu\text{m}$.

A low noise f.e.t. pre-amplifier was used in conjunction with a main amplifier with passive shaping, a biased amplifier and a 400 channel pulse height analyser. The complete analysis system is shown in fig. 2 and includes a digital gain stabiliser which was necessary to prevent gain drift due to temperature or

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Fig. 1. Schematic diagram of the experimental chamber.

Fig. 2. Pulse analysis system.

other ambient variations. A normalised mercury wetted relay pulser was used to calibrate the multi-channel analyser in terms of energy and was additionally used as a source of reference pulses for the spectrum stabiliser.

3. Results

The detector system was optimised by selecting suitable pulse shaping in the main amplifier and choosing a source-detector distance for best resolution consistent with an acceptable count rate. A typical ^{241}Am alpha particle spectrum is shown in fig. 3 from which the total fwhm resolution, η , was found after a long count time to be 14 keV for the 5.486 MeV alpha peak. It was estimated that the window thickness of gold (about $40 \mu\text{g cm}^{-2}$) on the surface barrier detector would represent an energy loss of between 10 and 30 keV depending on incident energy. The additional energy straggling in this thickness of gold would be negligible within the limits of the present work. The total peak broadening (η), is comprised of components due to the detector (η_D), electronics (η_E), source (η_S), and absorber (η_A), from which

$$\eta^2 = \eta_D^2 + \eta_E^2 + \eta_S^2 + \eta_A^2.$$

Fig. 3. A typical ^{241}Am alpha particle spectrum.

Peak broadening due to the system must be subtracted from the total in order to measure energy straggling due to absorbing material, only, but η_D , η_E and η_S contribute negligible straggling for thick absorbers.

Although η is a satisfactory parameter describing the straggling of mono-energetic alpha particles, it may be preferable to define a new parameter which is the half-width at half the maximum on the high energy side of the peak, $\eta_{\frac{1}{2}H}$. This parameter may be of particular significance in studies using ^{241}Am where a second peak due to 12% of the total alpha emission rate occurs at 5.443 MeV as shown in fig. 3. Additionally, at very low energies, spectra tend to become merged with noise on the low energy side of a peak.

3.1. ABSORBER THICKNESS MEASUREMENT

Two methods were used to determine the thickness of the aluminium absorbers used in the experiment.

In the first method, the weight of the absorber was divided by the surface area and gave an accuracy within $\pm 2\%$. A Cahn electrobalance capable of measuring to $\pm 1 \mu\text{g}$ was used to weight the absorbers.

Fig. 4. Residual energy as a function of absorber thickness.

Fig. 5. Typical alpha particle spectra following partial absorption through aluminium absorbers.

which were circular with a diameter of 2.54 cm. A range of absorbers between 0.9 and 1.4 mg cm^{-2} were measured in this way.

A second method of determining absorber thickness was one in which numbers of absorbers of about 0.2 mg cm^{-2} were used to reduce the energy of 5.486 MeV alpha particles. The observed energy loss was used in conjunction with known stopping power data at that energy¹⁾ to determine absorber thickness. Fig. 4 shows the results from both experiments in terms of residual energy against absorber thickness.

3.2. STRAGGLING MEASUREMENTS

Energy spectra for a number of absorber thicknesses between 0.2 and 6.2 mg cm^{-2} were recorded and some typical spectra with absorber thickness Δx , most probable energy E , and straggling parameter η , are shown in fig. 5. Although all spectra are shown on the same energy scale, low energy straggling was measured using a biased amplifier.

Table 1 shows straggling parameters η and $\eta_{\frac{1}{2}H}$ as a function of absorber thickness. It may be noted that at high values of absorber thickness, the value of η cannot be determined because of the merging of noise pulses with those due to alpha particles, although cooling of the detector and the use of lower noise electronics may result in an improvement.

The straggling parameter η is shown in figs. 6 and 7 as a function of absorber thickness and energy loss respectively, while $\eta_{\frac{1}{2}H}$ is shown in fig. 8 as a function of absorber thickness.

TABLE I
Experimental data of the energy lost, residual energy, and straggling parameters for alpha particles passing through aluminium absorbers.

Absorber thickness (mg cm^{-2})	Energy loss (keV)	Residual energy (keV)	η (keV)	$\eta_{\frac{1}{2}H}$ (keV)
0.2	117	5369	43	24
0.4	215	5271	55	24
0.6	335	5151	69	29
0.9	534	4952	97	48
1.0	590	4896	97	38
1.4	832	4654	147	72
2.0	1274	4212	176	87
2.7	1767	3719	218	109
3.0	2009	3477	196	92
3.6	2449	3037	261	114
4.0	2726	2760	300	130
4.2	2973	2513	306	108
4.4	3297	2189	360	162
4.5	3235	2251	365	183
4.6	3532	1954	396	180
5.0	3784	1703	434	189
5.2	4011	1576	429	210
5.4	4285	1202	512	222
5.6	4679	808	649	273
5.7	4916	570	729	318
5.8	5034	452	682	320
5.9	4912	574	614	256
6.0	5103	383	542	328
6.1	5237	249	—	166
6.2	5252	234	—	150

Fig. 6. Energy straggling parameter η as a function of absorber thickness.

Fig. 7. Energy straggling parameter η as a function of energy loss.

4. Discussion

The accuracy of the technique was limited by the difficulty in obtaining very thin uniform absorbers of known thickness.

Of particular interest is the existence of a maximum straggling value and a region of sharply decreasing straggling that follows. A similar phenomenon has been observed by Rotundi and Geiger²) after a detailed analysis of the stopping of polonium alpha particles in air, but has not been previously reported in solids.

An explanation of the decreased straggling at low energies is suggested by reference to the stopping

Fig. 8. Energy straggling parameter $\eta_{1/2H}$ as a function of absorber thickness.

Fig. 9. Stopping power data for alpha particles in aluminium below 1.6 MeV. (● Chu et al.⁵); Δ Porot et al.⁶); $\circ$ Williamson et al.⁷); + Northcliffe et al.⁸)).

powers at these energies. Fig. 9 shows theoretical and experimental stopping power data for alpha particles in aluminium at energies below 1.6 MeV. It should be noted that the data shows maximum values of stopping power within the region of 500 keV to 600 keV, and a region of decreasing stopping power at energies below the maximum. If the energies of a spectrum of alpha particles incident on an absorber fall within the latter region, then the high and low energy ends of the distribution would be subject to high and low stopping powers respectively (i.e. high and low energy losses in the absorber). This results in an energy spectrum with a decrease in energy straggling – the “energy bunching” effect. The validity of this hypothesis has been tested by recording the following spectra as shown in fig. 10.

1. An energy spectrum with a most probable energy of about 400 keV and $\eta_{1/2H} = 300$ keV (curve A).
2. The resulting experimental energy spectrum after transmission through an additional 0.2 mg cm^{-2} of aluminium (curve B).

Fig. 10. Energy spectra with experimental and predicted energy straggling.

3. A predicted spectrum (curve C) obtained from spectrum A for an additional absorber thickness of 0.2 mg cm^{-2} . The peak energies of A and B were used to obtain a value of the stopping power for this absorber thickness and energy and, with the recent theoretical stopping powers of Northcliffe and Schilling⁸), the appropriate energy loss was calculated for each energy "window" as defined by the channel number on the multichannel analyser. The small amount of straggling introduced within each window has not been considered here.

The value of the straggling parameter, η_{H} , shows good agreement between the experimental observations of 170 keV and the predicted value of 200 keV, giving weight to the hypothesis that the "energy bunching" effect is a function of stopping power at low energies.

The work of Comfort¹) shows that straggling increases towards the end of the alpha particle range, but his choice of absorber thickness did not extend to values at which decreasing straggling could be observed. Although Comfort used 8.78 MeV alpha particles from ThC', his values for straggling are comparable with those in the present work, using 5.5 MeV alpha particles, and also show the marked trend towards increasing straggling at low energies.

Ramirez³), using an alpha particle beam from an accelerator, has measured straggling for a number of gases, and their results tend to show a decreased straggling at low energies for argon, krypton and xenon which they consider to be in conflict with Comfort's observations of increasing straggling. However, in general, the studies of Ramirez have been carried out under conditions where the decreasing stopping power may be more significant than in Comfort's work, but too few experimental results are given to allow a satisfactory analysis. The results of

both Comfort and Ramirez are consistent with the present work.

5. Conclusion

Energy straggling of alpha particles in aluminium has been shown to exhibit a region of rapid increase followed by a rapid decrease towards the end of the alpha particle range. The resulting "energy bunching" effect has been explained on the basis of stopping power data for alpha particles in aluminium at low energies.

This phenomenon, dependent on the decrease of stopping power with decreasing energy at low energies, will occur for all heavy charged particles independent of the physical phase of the absorber.

Further experimental work is presently being carried out with other absorbers and incident charged particles.

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