

The connection of a He-jet
recoil transport system to an ion source

by

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I. INTRODUCTION

A. Study of Unstable Nuclei

Interest in the properties of nuclei far from the line of β stability has grown considerably in recent years. Only lately have many of these nuclei been made accessible for study by the advent of such technologically advanced production processes as heavy-ion-induced reactions and high energy reactions [1-3]. Other β unstable nuclei, the products of fission, have been available for more than a generation, but nevertheless remain unexplored. The study of neutron-rich fission product activities is the focus of nuclear research at the Ames Laboratory Research Reactor, and the desire to exploit the total range of activities produced in fission motivates the program of development outlined below.

The justifications for studying very neutron-rich (or deficient) species are abundant [4-6]. A complete nuclear structure theory requires information (out to the limits of particle stability) on nuclidic masses, decay schemes, half-lives, magic regions, deformation regions, transition regions, etc. Examination of recently identified prompt and delayed particle emitters should yield a clearer conception of nuclear wave functions and allow further refinement of the mass formulas. The verification and extension of current astrophysical theories (the r -process, for example) depends in part upon our knowledge of nuclei lying far from β stability. In a more practical vein, knowledge of such information as delayed neutron properties and absolute γ - and β -ray

decay rates for fission product activities can be applied to nuclear power reactor technology.

Attempts to fill in the highly β unstable portions of the chart of nuclides using standard techniques have been frustrated by such difficulties as low yields, extraneous reaction products, and short half-lives. Consequently researchers of these frontier regions must improve existing techniques or pioneer new ones to overcome these difficulties [7, 8].

Most highly β unstable nuclides are produced with very low reaction yields. Therefore the obvious challenge initially is the production of these exotic nuclides in sufficient quantities for study. This is not as formidable a task today as it was in the past. With judicious choices of projectile, energy, and target a large variety of previously unexamined nuclei can be generated with reasonable activities at modern facilities [9]. It is of interest to note that the most copious and efficient method for producing nuclides on the neutron-rich side of β stability is also the oldest - thermal fission of ^{235}U .

Another production-related problem is the lack of chemical and/or isotopic specificity in reaction products. Regardless of the method used to furnish spectroscopists with a given nuclide, the large distribution of nuclei that are inevitably formed with it make immediate detailed scrutiny difficult. Thus it is often necessary to use a mass separating system to avoid the difficulties of identifying the decay of interest in a complex mixture of nuclei.

The isolation and study of nuclides becomes increasingly difficult with distance from the stability line because the half-lives decrease rapidly to seconds or less. Since, due to counting limitations, it is usually impossible to obtain statistically significant information during a single half-life or less, an on-line procedure is indispensable. In some instances (especially fission), the required information for a given nuclide can be masked by the presence of its precursor or its daughters. Thus an arrangement which removes or suppresses daughter product interference renders an isotope separation on-line even more useful.

Very generally, an isotope separator on-line (ISOL) system is one in which a mass separating device is used in conjunction with an activity-producing particle flux to supply spectroscopic instruments with constantly renewed, isotopically or isobarically pure sources of short-lived nuclides. A concise presentation of the ISOL concept and of several ISOL configurations classified as to type of activity transport and mass analyzer is given in reference [8]. Since excellent surveys of ISOL systems are available [10], further review will not be attempted here. Attention will be narrowed instead to the TRISTAN on-line isotope separator located at this laboratory.

B. TRISTAN

1. Capabilities

Details of the TRISTAN system are given elsewhere [11], so only a brief overview pertinent to this treatise is presented here. In its

present configuration,¹ shown in Fig. 1, an 8 g uranyl stearate target (containing 25% fully-enriched ^{235}U) is positioned in an external neutron flux of 4×10^9 n/cm²-sec (maximum), of which 65% are thermalized. The gaseous fission products Kr and Xe are released from the target and are transported in a time of approximately one second to the ion source through a teflon tube of 140 cm length and 12.5 mm inner diameter. This transport, which occurs in a stream of He (with a few percent Xe and Kr added), is considerably more rapid than in the 5.5 m long transport line of TRISTAN's original (1966) configuration, which took 18 sec. The main purpose of the He-Xe-Kr gas mixture is to support the plasma discharge in the ion source, since the fission product activity alone is insufficient to sustain the plasma.

The TRISTAN ion source, which is of the Nielsen or oscillating-electron type [12], precedes a conventional Scandinavian-type isotope separator. The separator is a 90° sector magnet of mean radius 160 cm, and has a mass range at the focal plane of $\pm 7\%$ of the central mass. A digital mass meter positively identifies the isobaric decay chain under study. Ion beam scanning and position stabilization capabilities permit separator operation with individual beam currents of the order of tens of pA.

Beyond the focal plane of the isotope separator a secondary beam handling device is employed. This device, consisting of a switching

¹At this writing a new arrangement, nicknamed TRISTAN II, is in the early phases of operation. TRISTAN II utilizes an in-beam ion source with modified ion optics, and should not be confused with the system outlined above.

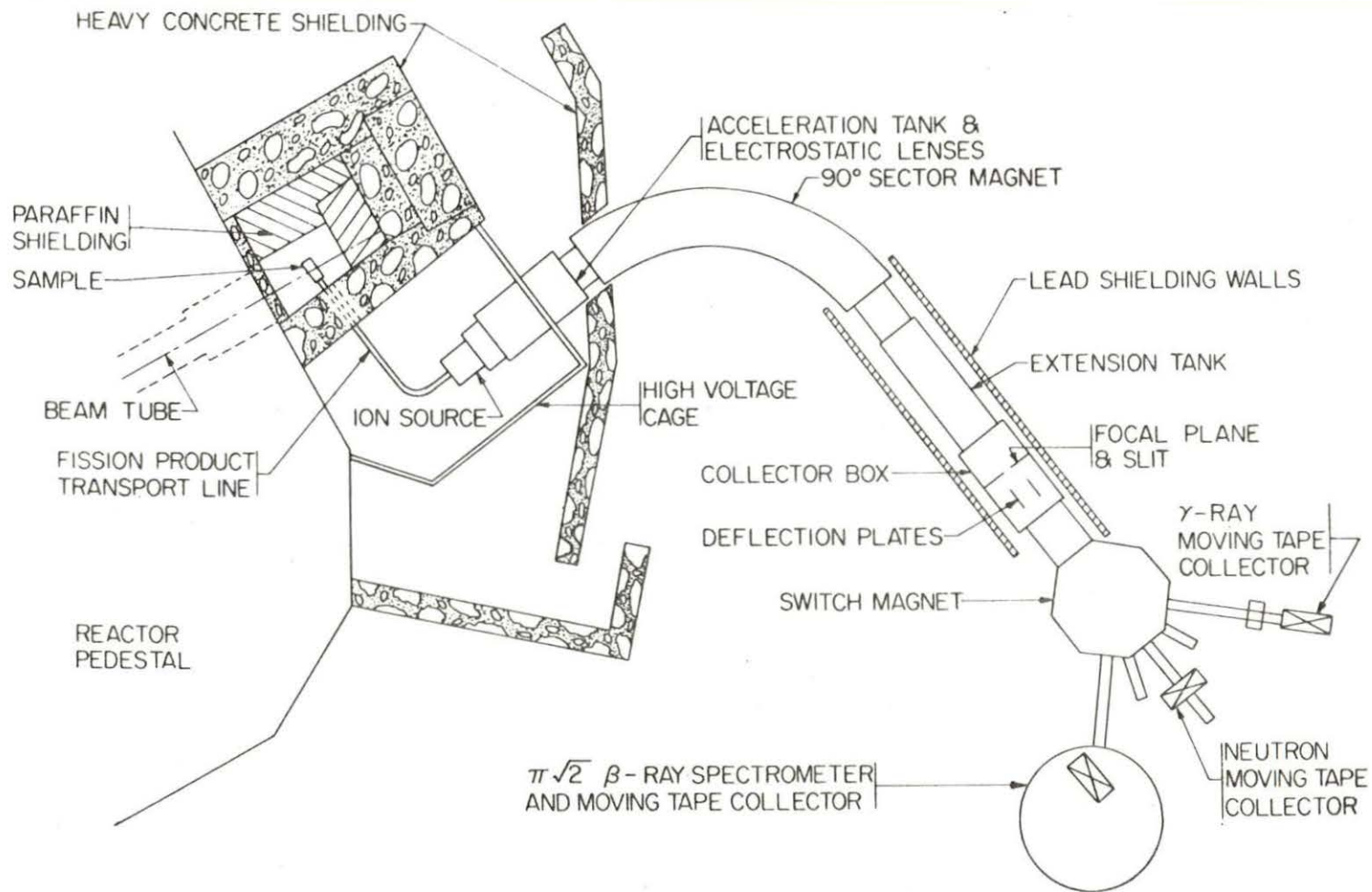


Fig. 1. Layout of TRISTAN on-line isotope separator system.

magnet and a vertical compression lens, provides a second stage of separation and refocuses the beam radially to one of three detection stations. Thus a single mass-selected ion beam can be directed away from the high background region of the separator collector into any one of the detection stations without disturbing the detector arrangements at the stations not in use. The switching magnet beam transport reduces cross-contamination resulting from gas scattering or charge exchange processes to non-detectable proportions.

At the central detection station an additional lens provides adequate focusing for delayed neutron studies [13]. Either a long counter or a ^3He neutron spectrometer can be accommodated here for delayed neutron multiscaling or spectrum measurements.

A $\pi\sqrt{2}$ double-focusing β -ray spectrometer with a beam stabilization device is connected to one of the 45° switch magnet ports [14]. This instrument is capable of providing accurate spectrum measurements with a resolution sufficient to resolve the numerous conversion electron peaks manifested in the decay of short-lived odd-A or odd-odd nuclei.

The other 45° port leads to a moving tape collector system used to separate the activities of long decay chains. The beam is deposited on a tape which may be moved past detectors at a continuous rate to provide parent activity enhancement, or in a stepped mode to enhance daughter activities. Thus, while the isotope separator provides an isotopically pure ion beam for collection, the moving tape

collector provides enhancement of the collected parent and subsequent daughter activities.

2. Limitations

The TRISTAN system, with its provisions for continuous, long-term experiments and its sophisticated beam handling capabilities, has proved to be a superior and versatile tool for the investigation of highly β -unstable nuclides in the neutron-rich region. However only partial acclaim should be acknowledged until TRISTAN can make available a wider range of short-lived mass-separated fission products. Throughout its nearly ten-year period of operation, studies have been limited to neutron-rich nuclei populated in the decays of the gaseous fission products krypton and xenon and their short-lived daughter activities.

The restriction of TRISTAN's productivity to rare gas activities is a consequence of the temperature and pressure in the target chamber and transport line. The target is unheated, so only those species with sufficiently high volatility at ambient temperature, which is at most a few degrees above room temperature, will diffuse from the target material. Since none of the fission products except krypton and xenon is gaseous at this temperature, removal of the non-volatile elements from the target material by diffusion is impossible without heating the target to several hundred degrees. The only alternative is to use a carrier gas to stop the non-volatile species as they recoil from fission events and then to sweep them from the target.

As stated earlier, a certain amount of support gas is required to sustain the ion source plasma discharge. Unfortunately the support gas functions ineffectively as a carrier gas at the very low pressures necessary for efficient operation of the ion source. At low pressures the mean free paths and recoil ranges of the fission products are large compared to the transport line radius and the target dimensions, and the flow mechanism is dominated by molecular collisions with the walls (molecular flow), rather than by intermolecular interaction (viscous flow). Profoundly affecting the transmission efficiency for a fission product, therefore, is its sticking coefficient, or probability per collision with the walls that it does not re-enter the gas phase [15]. Noble gases, because of their high volatility and low chemical reactivity, have very small sticking coefficients and can therefore be transmitted in a molecular flow with reasonable efficiency. The non-gaseous fission products have small or zero transmission efficiencies because their sticking coefficients are much larger.

C. The HeJRT Method

For the past two years a developmental program has been sustained at this laboratory in an effort to acquire new approaches to provide on-line activities of non-gaseous fission products [16]. With the goal of increasing the speed and efficiency of transport to the ion source by reducing wall collisions, free path lengths, and recoil ranges, a study of the helium-jet recoil transport technique has been

made. Presented in this thesis is a review of that study, with primary emphasis given to the design and testing of an arrangement in which a He-jet recoil transport (HeJRT) system is connected to a TRISTAN ion source.

1. Perspective

In one of the HeJRT method forerunners, the system used by Ghiorso in the identification of element 102 [17], recoil ions thermalized in He were transported to a collector using electrostatic fields. A few years later differential pumping, instead of electrostatic fields, was introduced to effect the transport [18, 19]. Thermalization and transport are still the primary elements of the HeJRT method today. The recoil atoms from nuclear reactions are either thermalized in the target and diffuse from it into the carrier gas or are ejected from the target into the thermalizing (carrier) gas by the primary momentum imparted during the nuclear reaction itself. The carrier gas containing the thermalized reaction products is then transferred from the target chamber by differential pumping through an orifice or a capillary to a separate chamber for detection or further handling. The speed and efficiency of the transport process is determined by the mass throughput, which is a function of the sticking coefficients of the activities being transported and system characteristics such as thermalization chamber pressure and volume, capillary length and cross-section, and pumping capacity.

The HeJRT method appears to be the most versatile gas transport technique in wide use today. Since its accidental discovery in 1961 by Macfarlane and Griffioen [in 20], the application of the HeJRT method to nuclear research has received much more attention than has the systematic, thorough understanding of the method itself. An understanding of the method would require extensive excursions into such specialized fields as plasmas, surface chemistry, fluid dynamics, and macromolecular phenomena - an expenditure of time and effort few active nuclear scientists are able to make.

In spite of the general lack of understanding of many of the mechanisms involved in the HeJRT method, it is empirically a fairly straightforward technique. The essential independence of chemical properties, combined with a capability of giving highly efficient, very rapid transport over relatively long distances make the HeJRT method particularly suitable for the on-line handling and study of short-lived radioactive nuclides. The HeJRT method has been used extensively to study heavy elements at Berkeley and Dubna [21-24]. At accelerator facilities it has been successfully employed to remove ultra neutron-deficient α - and p-emitting nuclides with half-lives in the msec range to low-background counting areas [20, 25-35]. Similarly the method has facilitated the β and γ spectroscopy of short-lived reaction recoils [36-38] and fission products from both thermal [39] and fast [40-42] neutron-induced fissions as well as spontaneous sources [43]. Recoils have even been transported via the He-jet into the liquid phase for fast radiochemical separations [44-47].

Recent efforts to connect a HeJRT system to the ion source of a mass separator have met with very limited success [39, 42, 48]. The reasons are not clear. The combined system makes no apparent pumping or electrical demands that cannot be met with standard equipment. Large pressure and temperature gradients present rigorous technical challenges, but to imaginative designers and skilled machinists such problems are not insurmountable. The root of the difficulties is still in the lack of understanding of the fine details of the mechanisms in the HeJRT method and their dependence on various physical parameters. The absence of these insights makes it difficult to predict the behavior of a He-jet-ion source system before assembly. It is appropriate, nevertheless, to preface our experimental observations with a brief discussion of the existing rudimentary theories and empirical observations regarding thermalization and transport in the HeJRT method.

2. Thermalization

In the presence of a thermalizing medium such as helium, fission products emerging from the target material rapidly lose energy and capture electrons until they become thermalized and attain a charge state determined by their ionization potentials relative to the ionization potential of helium [49]. In order to minimize losses the volume and pressure of the helium should be such that the non-volatile recoils are stopped before colliding with the walls. Clearly the thermalizing volume should be as small as possible to minimize evacuation time. The

pressure should be high enough to assure complete stopping of the fission products, but not so high that excessive turbulence results. These notions have been confirmed experimentally by Macfarlane [27].

The flow of gas in the target chamber is complex. Mild turbulent flow dominates in the target, and a vortex is probably formed at the capillary entrance. The recoils are caught up in the vortex and, barring collisions with the walls, they will enter the capillary, where laminar flow is established within a short distance from the inlet (depending upon the Reynolds number of the flow; see section on flow). If the gas pressure increases, the vortex extends toward the target walls and eventually interacts with them depositing a fraction of the activity on the surface.

The nature of the junction of the target chamber to the capillary has been found to have a marked effect on both efficiency and speed [27]. Best results are obtained when the capillary is extended about 2 mm into the target chamber, rather than in a flush or tapered joint. This minimizes turbulence by keeping the vortex away from the container surfaces and allowing the gas to set up its own natural flow lines into the capillary. In addition the re-entrant geometry provides optimal convergence of the gas jet to laminar flow [50].

3. Transport

a. Clusters In observing the free-expanding jet emerging from the low-pressure (20 μm) end of a capillary, Jungclas, et al. [20] noted a much smaller angular divergence (0.5°) than expected for the mass-8

(^8Li) beam of reaction recoils carried by the helium [51]. The mass and charge distributions of the heavier component of the beam were analyzed by injecting it through an orifice and into another vacuum chamber in which an electric field was defined by parallel plates. The beam was found to be comprised of 25% positive ions, 16% negative ions, and a 59% neutral component, and the masses were 10^7 to 10^8 u. The conclusion was that the reaction recoils became attached to large molecular clusters produced from impurities in the helium by the intense ionization generated as the cyclotron beam passed through the target chamber.

It is now known that ultrapure He produces less efficient transport than does impure He [20, 52-55]. The importance of cluster molecules was not fully realized until typical transport lengths in HeJRT systems increased to present size. Efficient transport with pure He is limited to between 50 cm and 100 cm in most systems [53], but the use of clusters extends this limit to as high as 200 m [56]. The nature of these clusters, the nature of the bonding between the radioactivity and the clusters, the role of impurities in a HeJRT system and the relationship between these impurities and chemical selectivity of the transport process are areas which are not well understood.

There is evidence that prior to the attachment of the reaction recoils to the impurities, the latter undergo radiation-chemistry break-up. This was clearly demonstrated in experiments that attempted to transport thermalized fission fragments from a spontaneous source

(^{252}Cf) using the HeJRT method [42, 43]. Infinitesimal transport yields were obtained until the He cell was irradiated with intense ultraviolet light; the yield then increased dramatically. Thus electromagnetic radiation was shown to be a necessary stimulus to the formation of clusters from the additives in the He. Most evidence suggests that the clusters are condensation aerosol particles formed when the radiation generates the breakup of impurity vapors [29, 43, 56-60].

A fundamental unanswered question concerns the nature of the mechanism through which a recoil becomes bound to a cluster. A number of descriptions of the attachment mechanism have been proposed. If the clusters form in the presence of the recoil ions, then entrapment through nucleation [61] or polymerization is plausible. On the other hand, the binding may be a surface interaction such as chemisorption or physical adsorption of the activities onto the cluster [62]. Based on the experiences of Macfarlane [38] in depositing activities from a He-jet on a heated catcher foil, and considering the relative success of Kosanke [45] and Aumann [46] in achieving fast liquid radiochemical separations, the cluster-recoil binding is probably best understood in terms of a weak van der Waals or surface effect.

It is known that small ions became attached to large charged and neutral condensation aerosol particles by forces that are presumed to be surface and van der Waals in nature. The number of small ions n that become attached to aerosol particles carried in a flowing gas is given by [63]

$$\eta = \eta_0 [1 - \exp(-v\phi)]$$

where

η_0 = the original number of ions,

ϕ = the flow rate of the carrier gas,

v = a parameter related to the sticking coefficient between the aerosol and the small ion as well as to the concentration of aerosols in the carrier gas.

This relationship has been verified experimentally using ethylene clusters (aerosol particles) to transport fission products (small ions) in a HeJRT system [58].

The connection between transport efficiency and impurity concentration has resulted in considerable efforts to find an ideal additive for the He that will result in the highest efficiency [29, 42, 54, 57, 64]. It is now believed that the size and not the content of the clusters is important in determining the relative transport efficiency, and furthermore that the clusters are, with but a few exceptions, chemically non-selective [64]. Nevertheless, a very slight Z-dependence has been observed in the transport efficiency of fission products [42]. This may be due to the various ranges and effective charges possessed by the recoils in the thermalization chamber.

Reductions in transport efficiency occur when the clusters contact the capillary walls and the activities are adsorbed thereon. In an

excellent discussion by Wieseahn et al. [58] of losses in the capillary, semi-empirical equations that describe sedimentation (gravitation) and Brownian motion [65] are applied to capillaries with various dimensions and flows. It is shown that most capillaries act as filters which remove large clusters by sedimentation and small ones by Brownian motion, while particles with radii of greater than about $0.01 \mu\text{m}$ and less than about $2 \mu\text{m}$ are transmitted without loss.¹ It is also shown that, due to coagulation effects [66], the cluster size distribution will get narrower. This means that in higher cluster concentrations, small particles will be absorbed by larger ones. This loss probably dominates over the loss of small clusters due to Brownian motion and sets a lower limit on the useful cluster size.

b. Dynamics The theoretical description of the flow of a homogeneous gas in a capillary tube is not new or difficult. In a HeJRT system in which clusters are used, however, the effects of the impurity in the He on the gas dynamics is justifiably a matter of concern. With specific regard to the transit time, the pertinent questions are (1) whether the clusters move with the same speed as the He and (2) whether the presence of clusters in the He causes a reduction of the maximum attainable speed of the gas.

Relative to He, the clusters are very large and massive particles, so it is reasonable to suspect a certain amount of "slip" between the

¹This would explain why pure He is capable of very limited transport of bare recoils, which have diameters in the range $5 \times 10^{-4} \mu\text{m}$ to $10^{-3} \mu\text{m}$.

two species. The velocity gradient of a cluster in the flow direction is given by [67]

$$\frac{dV_c}{dx} = \frac{9\eta}{2\rho_c r^2} \left(\frac{V}{V_c} - 1 \right)$$

where

r = the radius of the cluster,

V_c = the speed of the cluster,

V = the speed of the carrier gas,

ρ_c = the cluster density,

η = the viscosity of the carrier gas.

Assuming a typical maximum value for dV/dx of 10^4 s^{-1} [58, 67] and a cluster radius of $0.5 \mu\text{m}$, one can obtain for the quantity in the brackets values of less than 3×10^{-2} for most hydrocarbon clusters in He. With but a 3% difference between the cluster and gas speeds, only negligible differences in the transit times would result.

It has been predicted [68] that the speed of sound of the He will be reduced by the addition of clusters to the flow. This reduction is given by

$$\left(V_s' / V_s \right)^2 = \frac{1 + \alpha\xi}{(1 - \epsilon)^2 (1 + \alpha) (1 + \gamma\alpha\xi)}$$

where

V_s' = the speed of sound of the mixture,

V_s = the speed of sound of the carrier,

ϵ = the volume fraction of the clusters,

α = the mass-flow ratio (loading ratio) = $\frac{\phi}{1-\phi} \frac{V_c}{V}$,

ϕ = the mass fraction of clusters = $\sigma_c(\sigma + \sigma_c)^{-1}$,

ρ = the carrier gas density,

σ = the partial carrier gas density = $(1 - \epsilon)\rho$,

σ_c = the partial cluster density = $\epsilon\rho_c$.

ξ = c/c_p with c = specific heat of clusters and c_p = constant pressure specific heat of carrier gas,

γ = the ratio of specific heats c_p/c_v for the carrier gas.

If hydrocarbon clusters of radius 0.5 μm are again considered, then with a number density of 10^6 cm^{-3} , the calculated reduction of the speed of sound is less than 1%.

Because both the effects discussed above are so small, they will be neglected in the following development; instead the flow will be treated as that of a homogeneous gas. Furthermore, since He is so nearly a perfect gas, ideal gas laws will be freely utilized.

Assuming adiabatic flow of the gas in a tube of constant circular cross-section and that frictional forces arise from the viscosity of the fluid, the one-dimensional theoretical result from gas dynamics is that the gas accelerates down the tube with the local velocity given by [40, 46, 54, 69, 70]

$$\frac{x}{D} = \frac{1}{f} \left[\ln\left(\frac{V_1}{V}\right)^{(\gamma+1)/\gamma} + \frac{T_1 R_0}{M} \left(\frac{1}{V_1^2} - \frac{1}{V^2} \right) \right] \quad (1)$$

where

x = the position along the capillary,

V_1 = the speed in the capillary entrance,

V = the speed at the position x ,

D = the capillary diameter,

T_1 = the gas temperature in the capillary entrance,

R_0 = the universal gas constant,

M = the molecular weight of the gas,

f = the coefficient of flow resistance.

The coefficient of resistance (f) is a function of the Reynolds number, $\mathcal{R} = \rho V D / \eta$, where ρ and η are the gas density and viscosity, respectively. For laminar flow, occurring for $\mathcal{R} \lesssim 2300$ [46], the

coefficient of resistance is given by $f = 64/R$, while for turbulent flow, which occurs for $R > 2300$, $f = 0.3164/R^{1/4}$ [70].

Within the capillary the gas speed is everywhere subsonic, increasing to a maximum at the exit. When the pressure difference between the ends of the capillary is great enough, sonic velocity is achieved at the exit. When this condition, called choked flow, exists, no increase in the pressure differential will further increase the gas speed [71]. Sonic velocity in an ideal gas is given by [71]

$$V_s = (\gamma R_0 T/M)^{1/2}$$

where T is related to the local speed of the gas by

$$T = T_0 \left[1 - \frac{1}{2}(\gamma - 1) \frac{V^2}{V_s^2} \right] . \quad (2)$$

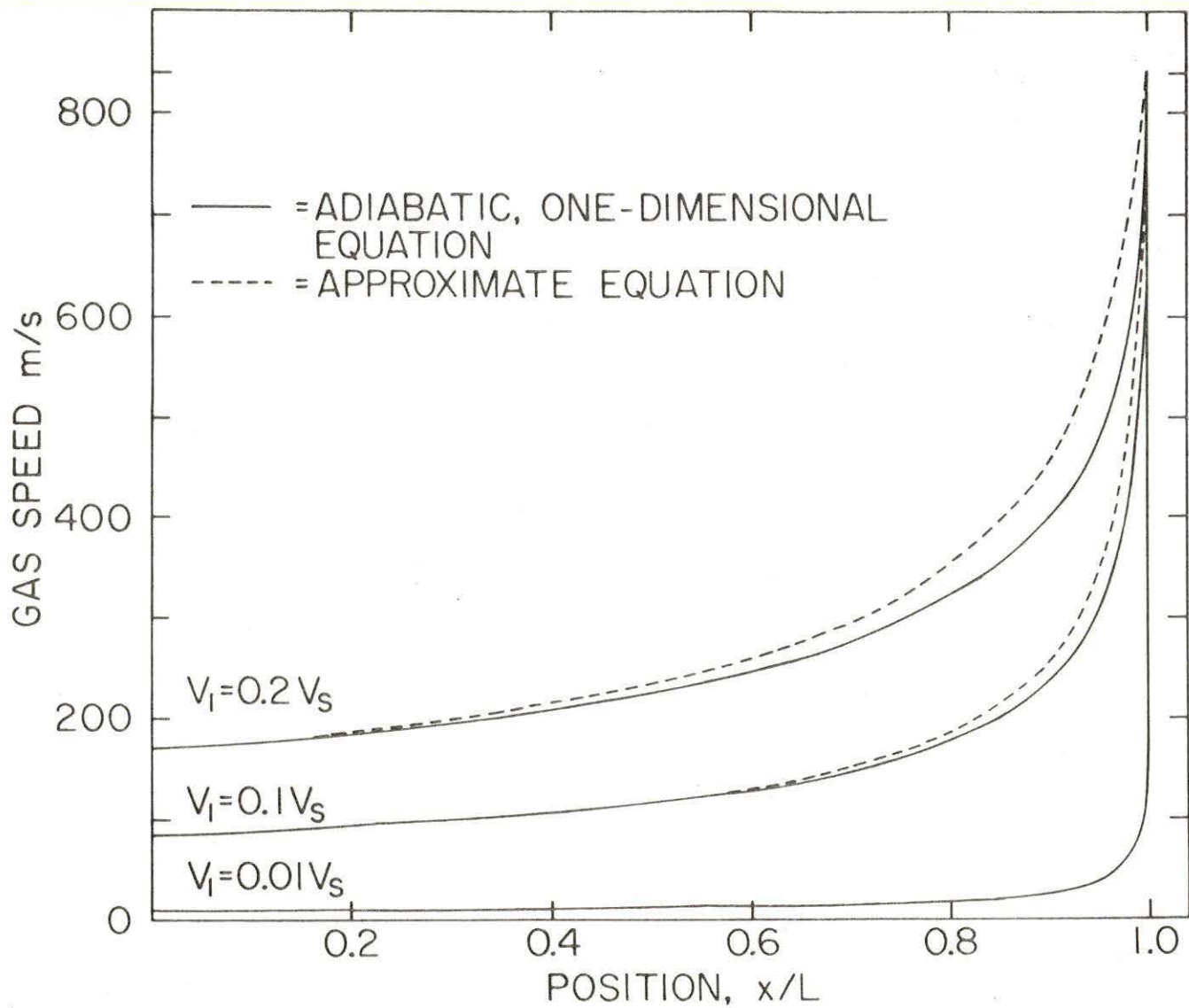
Thus the gas temperature at the capillary exit, where $V = V_s$, is

$$T_s = 2T_0/(\gamma + 1) .$$

For He at $T_0 = 20^\circ$ C, the sound velocity V_s is 970 m/s, while at the end of a capillary in choked flow, $V_s = 840$ m/s due to the temperature decrease.

From Eq. (1) the speed as a function of position in the capillary is plotted in Fig. 2 as solid lines for choked He flow with various inlet speeds. The speed is seen to increase slowly in the majority

Fig. 2. Plot of gas speed as a function of position in a capillary.



of the length of the capillary, then rapidly at the end. Clearly high inlet velocities are required to minimize capillary transit time.

In most HeJRT systems the inlet speed, which can be estimated from He flow rates [40, 53, 69] or measured elapsed times for activity transport [41, 56], is less than 85 m/s, or about one-tenth the speed of He in choked flow. The observation that, for $V_1/V_s \leq 0.1$, the logarithmic term in Eq. (1) is less than 5% of the other term for all V from V_1 to V_s suggests that a reasonable approximation should result when the term is neglected. It should be noted also that, in the case $V_1/V_s \leq 0.1$, there is little temperature drop ($< 1\%$) from T_0 to T_1 as the gas flows into the capillary entrance [see Eq. (2)]. A similar relationship holds for p_0 and p_1 [71].

Therefore it is found that at the end of a capillary in choked flow, with $T_1 = T_0$ and with the logarithmic term ignored, Eq. (1) becomes

$$\frac{L}{D} = \frac{T_0 R_0}{f^M} \left(\frac{1}{V_1^2} - \frac{1}{V_s^2} \right) . \quad (3)$$

Then by combining Eqs. (1) and (3) and dropping the logarithmic term, the simplified expression for speed versus position results:

$$\frac{x}{L} = \left(1 - \frac{V_1^2}{V^2} \right) \left(1 - \frac{V_1^2}{V_s^2} \right)^{-1} \quad (4)$$

where $V_s = V_s(T_s) = 840$ m/s. Using this equation, the speed is plotted as dashed lines in Fig. 2 for different values of V_1 . For $V_1 \leq 0.1 V_s$

the difference from the solid curves calculated with Eq. (1) is seen to be very slight.

The elapsed time for a gas molecule in the capillary is found by solving Eq. (4) for V and then integrating:

$$\Delta t = \frac{2L/3V_1}{1 - (V_1^2/V_s^2)} . \quad (5)$$

Equation (3) yields a simplified expression for V_1 upon substituting the relation $f = 64/\mathcal{R}$ in which the ideal gas relationship $\rho_0 = p_0^M/T_0R_0$ is used, and dropping the term $1/V_s^2$, which is less than 1% of $1/V_1^2$:

$$V_1 = p_0 D^2 / 64 \eta L . \quad (6)$$

Equation (6) also follows from the Hagan-Poiseuille law for gas flow in tubes [70, 72].

To optimize V_1 for a given length of capillary one can increase the pressure p_0 or diameter D , subject to the condition that laminar flow is maintained ($\mathcal{R} < 2300$). The expression of the Reynolds number in terms of V_1 ,

$$\mathcal{R} = \frac{64V_1^2 LM}{DR_0 T_0} \quad (7)$$

indicates that, for a given length, large-diameter capillaries permit higher values for V_1 before turbulence sets in. The dependence of the

Reynolds number on the molecular weight of the gas indicates light gases will permit the highest-speed non-turbulent flows.

The assumption has been made that the pressure in the chamber at the exit of the capillary has been held low enough that the gas speed reaches V_s at the exit. For an ideal gas it has been shown [70] that the exhaust pressure p_3 must be less than that given by

$$\frac{p_3}{p_0} = \frac{V_1}{V_s} \left[\frac{2 + (\gamma - 1)V_1^2/V_s^2}{\gamma + 1} \right]^{\frac{1}{2}}$$

for choked flow to exit. In the range of inlet speeds for which Eqs. (3)-(7) are good approximations, the choked flow condition reduces to

$$\frac{p_3}{p_0} = \frac{V_1}{V_s} \left(\frac{2}{\gamma + 1} \right)^{\frac{1}{2}} \quad (8)$$

The transmission efficiency in a cluster-loaded HeJRT system does not depend essentially on the length of the capillary at distances of several meters or even tens of meters [54]. Apparently this is due to the existence of a mechanism that forces the heavier constituents to the center of the capillary cross-section, reducing contact with the walls. Such behavior in laminar flows was predicted by Jeffery in 1922 and it has been observed by Becker and Bier [73], Jungclas et al. [20], and Wilhelm et al. [42]. When a regular laminar flow pattern has been established with the cluster concentrated near the axis of the tube, absorption onto the walls is negligible. Thus the problem of transporting activities over long distances appears to be in effectively bringing the

clusters to the center of the flow pattern and then in maintaining sufficiently laminar flow in the capillary.

Most serious losses due to diffusion are concentrated in the first few cm past the junction between the target chamber and the capillary [53]. Only in laminar flow is adsorption minimized by the focusing of the clusters near the axis. Over the entrance area of the capillary, however, the flow velocity is approximately uniform. As the gas moves down the tube, frictional forces continually modify the flow until, at a distance ℓ from the entrance, a parabolic (laminar) velocity profile is established which then remains constant throughout the remainder of the passage through the tube. The distance ℓ at which the flow is fully developed is given approximately by [72]

$$\ell = 0.227 a \Re \quad (9)$$

where a is the capillary radius in cm and \Re is the Reynolds number. Since it is desirable to establish fully developed flow in as short a distance as possible to reduce losses, the diameter or the Reynolds number (or both) should be minimized. This can only be accomplished at the cost of increased transit time arising from a reduced inlet speed [see Eqs. (5) and (7)]. For example, in order to reduce ℓ to under 1 cm in a 5 m-long capillary of diameter 1 mm, the Reynolds number must be less than 90. This limits the inlet speed to about 13 m/s and gives a transit time greater than 250 ms.

D. Connecting a He-Jet to an Ion Source

1. Perspective

HeJRT systems have been connected to mass separator systems with only limited success [40, 42, 48, 74]. During these early investigations it has become apparent that a number of complex technical problems are involved and that the understanding and optimization of several coupled parameters must be achieved. Unusual pressure and temperature differentials present the primary technical obstacles. The bulk flow of a HeJRT system and its relatively high exhaust pressure (typically a few hundred mTorr) are incompatible with the very low pressure (a few mTorr) of a separator ion source. The high-temperature (1500° - 2000° C) hostile environment of a typical ion source can cause pre-entrant vaporization of the clusters and loss of the activities before ionization can take place. Parameters such as target pressure, flow rate, ion source pressure, and beam current are inter-related, and all affect system efficiency. In short, a He-jet-ion source arrangement is a very complex and sensitive apparatus.

2. The skimmer

One of the crucial developments in achieving an operational system was the incorporation of an Abschäler [73], or skimming device [20, 29, 42, 55, 56]. The purpose of the skimmer is to separate the bulk of the He from the clusters before injecting the stream into the ion source. The ease with which this can be accomplished is an additional benefit of the large cluster masses. As stated earlier, the clusters concentrate

at the capillary axis because they experience no net radial force. This is not true of the He atoms which possess radial velocity components due to their thermal energy. As the jet emerges from the nozzle, or capillary tip, into the low-pressure exhaust chamber, He-He and He-cluster scattering occurs. In a collision the light He atoms are scattered through large angles while the clusters receive very small deflections and remain confined within a very small solid angle in the forward direction. A conical orifice coaxially placed the proper distance from the nozzle will transmit the clusters but not the He, so that the He can be pumped off. By using one or more such skimmers, the ion source can be maintained at the low pressure required for efficient operation. As would be expected from symmetry considerations, nozzle-skimmer alignment is critical for good skimming efficiency [29, 42].

II. EXPERIMENTAL ARRANGEMENT

Two years ago at this laboratory, John C. Pacer set up a HeJRT system, connected it to an ion source on a test bench, and initiated a program of testing and development. The original configuration, shown in Fig. 3, featured a standard HeJRT system with a 5 m long, 0.9 mm (i.d.) teflon capillary, a skimmer, and an oscillating-electron-type ion source. A large number of refinements (detailed below) of the ion source and skimmer have been made since this particular arrangement was used, but the He-jet and many of the optimum operating parameters determined by Pacer [16, 75] are presently intact.

A. The HeJRT System

1. The target chamber

The target chamber is positioned just inside the reactor face in a non-directional thermal neutron flux of $2 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$. It is a cylindrical Al can with a length of 3.5 cm and an inside diameter of 2 cm. Lining the can is a concentric cylinder of UO_2 -impregnated graphite cloth containing about 250 mg of ^{235}U . The 0.6 mm thick graphite cloth partially degrades the energy of fission recoils to permit attachment to the clusters within the 9 cm^3 target volume. A 10 cm length of 0.5 mm (i.d.) stainless steel capillary tubing protrudes about 3 mm into the chamber along its axis and is connected to the teflon capillary on the outside. In preliminary tests [16, 75] it was found that the highest target pressure at which the He-jet could be run

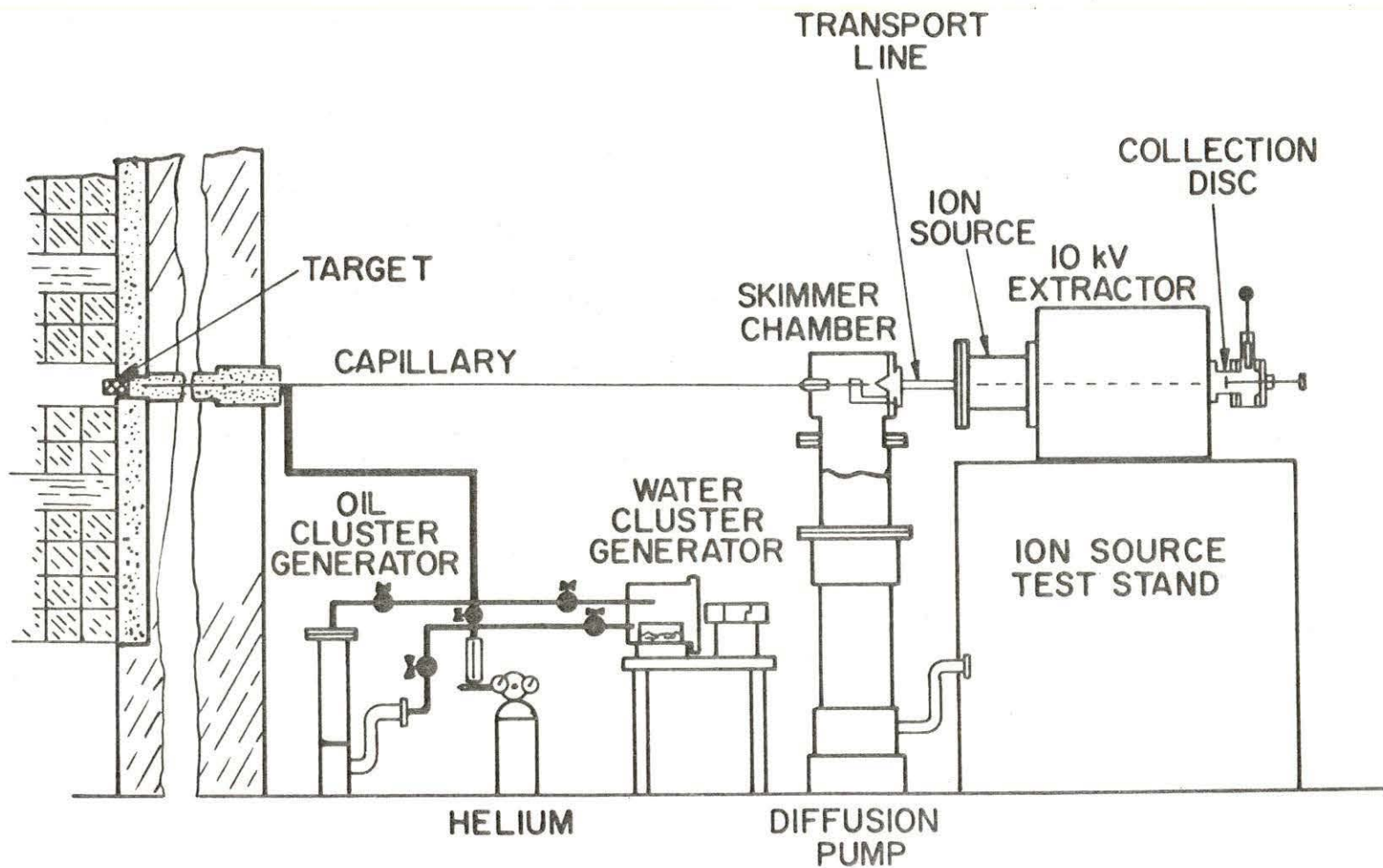


Fig. 3. Schematic diagram of the original He-jet-ion source assembly.

without erratic operation by the exhaust (skimmer) pump was about 520 Torr, which corresponds to a He flow rate of $2 \text{ cm}^3/\text{s}$. No reduction in beam current was observed when lower target pressures were tested, but proportionate reductions in the inlet speed should be expected at the lower target pressures [see Eq. (6)]. Therefore the target pressure is normally maintained at 520 Torr.

2. Cluster generation

Clusters generated by two different methods have been used with comparable transmission efficiency. As shown in Fig. 3, He entering the target chamber can be loaded with either oil or water clusters by adjusting the appropriate valves. Oil clusters are formed by heating diffusion pump oils (typically Dow Corning DC-704) in an old 2" diffusion pump. Water clusters are generated by irradiating the vapor above an open dish of water with a Hg arc discharge lamp. The oil clusters tended to coagulate to form large droplets on the walls of the capillary, so water clusters have been used almost exclusively in spite of their lower temperature for vaporization.

B. The Skimmer

The nozzle-skimmer is located inside a chamber which is mounted directly on a 6" diffusion pump (NRC Model VHS-6) with a pumping speed of 3000 l/s (He). The forepump, a Cenco Hyvac Model 150, has a pumping capacity of 960 l/m (34 CFM). Under best operating conditions the pressure in the skimmer chamber is about 400 μTorr . The alignment

between the nozzle and the 1.3 mm diameter skimmer cone hole was maintained at better than 0.02 mm with a one-piece holder. It was demonstrated [16, 75] that the nozzle-skimmer separation could be varied over a fairly wide range (4-9 mm) without seriously affecting the transmission efficiency, thereby signaling the apparent absence of the effects of shock wave phenomena observed by others at higher skimmer pressure [76]. Most subsequent work has been done with a nozzle-skimmer separation of 5.1 mm. Non-gaseous fission products were among those detected in an off-line analysis by Ge(Li) spectroscopy of a collection disk positioned after the skimmer [16, 75], and the transmission was determined to be satisfactory. Sufficient separation of the He from the jet was also obtained, since the pressure in the volume beyond the skimmer was less than 10 μ Torr.

C. He-Jet Ion Source Development

After it was demonstrated that the He-jet-skimmer system was functioning satisfactorily, the skimmer and a "stock" TRISTAN ion source [11] were coupled by a 25 cm stainless steel transfer tube. Originally the large diffusion pump on which the skimmer chamber was mounted rested on the floor, and the ion source, with its associated pumps and power supplies, was separately attached to a rolling test stand. In this configuration it was very difficult to maintain precise alignment from the skimmer orifice through the transfer tube and ion source to the ion source extractor 75 cm beyond the skimmer. This difficulty was minimized by integrating the skimmer pump and test stand into one unit.

The very high temperatures in the ion source presented a more serious problem. It was observed [16, 75] that in the transfer tube between the skimmer and ion source the clusters had acquired sufficient thermal energy to vaporize and deposit the non-gaseous activities on the walls of the tube. Unsuccessful attempts to prevent this premature cluster breakup were made with a number of devices [75]. Ultimately a new design of the ion source assembly was tested. In the original arrangement, the transfer tube was cooled by a low-volume water jacket to within about 1 cm from the ion source entry port. The cathode and anode electrical feed-throughs in the old arrangement projected axially from the ion source base toward the skimmer, prohibiting further reduction of the transfer tube length. The new design included higher-volume continuous-flow water cooling, an aluminum heat sink, and radial deployment of the electrical leads. This radial geometry permitted the elimination of the transfer tube and a reduction of the nozzle-to-plasma distance of about 25 cm. It was anticipated that the shorter transfer distance and the more efficient cooling arrangement would complement each other to reduce fission product loss in the transfer area.

With the new design, new problems were encountered. It was immediately evident that at least a fraction of the water clusters were not vaporizing prematurely because the ion source filament oxidized very rapidly. The tungsten oxides then sputtered onto the base insulator plate and shorted the filament to the anode, which should operate at a potential difference of about 50 volts. Filament lifetime was about

24 hours for tungsten and 1-4 hours for thoriated tungsten. After an exhaustive search for a solution to the problem it was concluded that no refractory material of sufficient chemical stability is available to supersede the tungsten. It was then discovered that thoroughly cleaning a tungsten filament with emery paper before installing it would curtail the sputtering and extend the filament lifetime considerably.

In the tests with this assembly, none of the activities deposited on a collection disk held at 10 kV after the ion source were determined to be non-gaseous in origin. Off-line Ge(Li) surveys of the individual components of the ion source revealed that a few non-volatile fission products (^{134}Te , ^{132}I , ^{134}I) had achieved entry into the ion source but had not been ionized and extracted. Since most of the activity seemed to be collected on the boron nitride components (base and end cap), it is possible that a chemical affinity of the boron nitride for the halogens exists. An alternative explanation for the holdup of non-volatile species in the ion source is that it was too cold, resulting in condensation of the activities on the surfaces. There was also a significant deposition of activity in the short tube between the skimmer and the ion source, indicating that the transport efficiency from the skimmer to the ion source was still not good.

With these results in mind, another modification to the system was made [16]. This design eliminated the short distance between the skimmer and the ion source by incorporating a water-cooled stainless

steel skimmer cone as the back of the ion source, similar to the approach by Mazumdar, et al. [74]. By placing the ion source as close as possible to the skimmer in this fashion, it was hoped that breakup of the clusters before entry to the ion source could be eliminated. Although there was a small yield of iodine (^{132}I and ^{134}I) with this configuration, no other non-gaseous activities were observed after the ion source. Upon dismantling the system, it was found that both Te and I activities were present at the boron nitride insulators inside the ion source.

Next, the base, anode, and end cap of the ion source were reconstructed of graphite to reduce the affinity of ion source materials for the fission products. Quartz rings insulated the anode from the base, and the water-cooled skimmer cone was unchanged. No increase in the amount of non-gaseous activities adsorbed on the collection disk was observed, but there was a deposit of such activities on the inside surface of the cone.

Since fission products will diffuse out of graphite at higher temperatures, it was proposed that lining the skimmer cone with a thin graphite cone would prevent condensation of the activities on the stainless steel cone. Accordingly a new graphite base was constructed similar to its predecessor, except that it incorporated a thin graphite cone. This left a narrow space of 3 mm between the cooled stainless cone and the hot graphite cone. Clusters surviving the graphite cone orifice were expected to break up inside the ion source and the activities deposited inside the ion source were expected to diffuse out

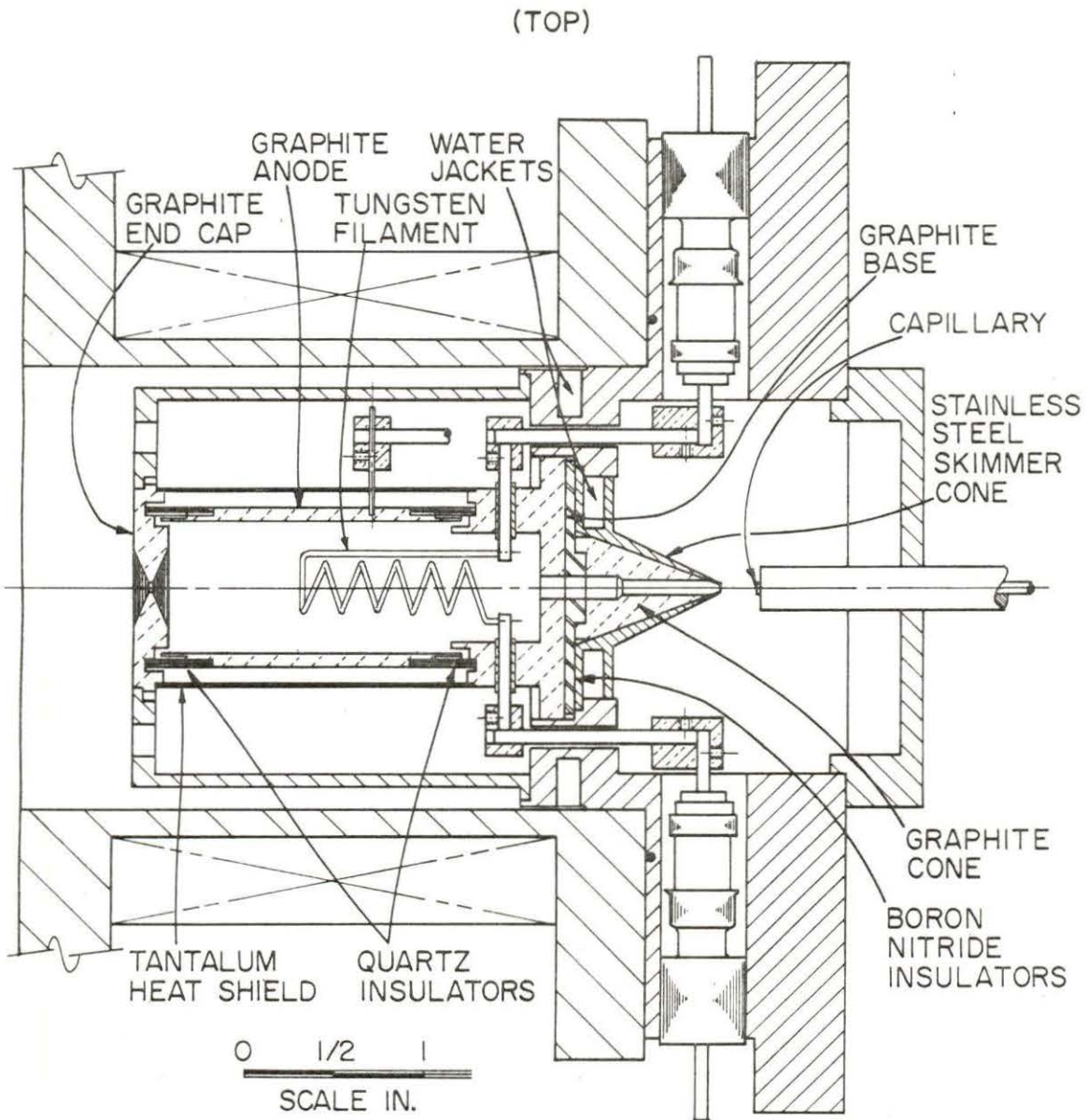
of the graphite and into the plasma discharge. Tests indicated that clusters broke up in the region between the two cones, again depositing the activities on the inside of the skimmer cone as before, instead of passing through the orifices as expected.

In the final arrangement, shown in Fig. 4, the first graphite base was reinstated with a separate graphite cone. This cone touches the stainless cone and is solid, with a narrow passage through its center and a flare at the exit end. A boron nitride disk heat-insulates the base from the cone. Recent tests of this arrangement still indicated no increase in the amount of non-gaseous activities collected after the ion source. However, the only measurable deposition of non-volatile activities is now on the inside surface of the end cap. This possibly indicates that all parts of the ion source except the end cap can be made hot enough to prevent condensation and adsorption of the non-gaseous fission products. Reduction of the thermal contact between the end cap and the aluminum ion source holder could cure the remaining problem.

End cap activities observed off-line by Ge(Li) spectroscopy are displayed in Figs. 5 and 6. Also shown are the gaseous activities observed on-line after the ion source and the fission products transmitted through the skimmer, as reported by Talbert et al. [16].

D. Summary of Performance

With a flow rate of $2 \text{ cm}^3/\text{s}$ and a thermalization volume of 9 cm^3 , an average target chamber sweep-out time of $\sim 4.5 \text{ s}$ might be expected.



He-JRT ION SOURCE ASSEMBLY

Fig. 4. Diagram of present He-JRT ion source assembly.

The inlet speed V_1 can be determined either of two ways. If the flow rate is divided by the entrance area of the capillary, an experimental value of V_1 is 1020 cm/s. If Eq. (6) is used, a theoretical value of 886 cm/s is obtained. The two results are in reasonable agreement considering (1) the He flow rate is measured before clusters are added, (2) the target pressure given is actually that of the supply line rather than that of the target volume itself, (3) the capillary cross-section is not constant, and (4) Eq. (6) is an approximation of an adiabatic one-dimensional relationship for an ideal gas. From Eq. (5) the capillary transit time for this system is about 0.4 s. Therefore, the expected total transport time for this HeJRT system is about 5 s.

Based on observations by Kirchner and Roeckl [77] of standard Nielsen ion sources and on the fact that the TRISTAN system is capable of providing activities with half-lives of less than a second [11], a delay time in the He-jet ion source of less than 1 s can reasonably be estimated.

In its present configuration, therefore, it is expected that the He-jet-ion source system should be able to provide activities with half-lives of greater than about 6 s. However, the shortest-lived species observed consistently are ^{90}Kr (32.3 s) and ^{139}Xe (39.7 s). No ^{91}Kr (8.57 s) has been detected, and ^{140}Xe (14.3 s) has appeared only in occasional runs. These results suggest that the effective target sweep-out time is at least 15 s. In the turbulence of the target chamber, perhaps the clusters do not respond as quickly as the He to the gentle pressure gradients present there.

In this system the ratio of skimmer pressure to target pressure is about 8×10^{-7} , which is much smaller than the maximum value of 9×10^{-3} given by Eq. (8) for choked flow. Thus it is relatively certain that the gas speed at the nozzle is sonic.

The Reynolds number for the flow in this system is about 47 [Eq. (7)], and Eq. (9) gives a distance of under 3 mm for fully developed flow at the inlet.

IV. CONCLUSIONS

Further improvement of both the ion source and the HeJRT components of the system could be made. As mentioned earlier, provisions for heating the end cap to higher temperatures should be made in an attempt to prevent deposit of non-volatile species before ionization.

More efficient transport and shorter transport times are desirable HeJRT system characteristics, but simultaneous achievement of the two should be very difficult. Increasing the target pressure, for instance, would shorten the transit time but increase inlet losses. Reducing the capillary length would improve both transport time and efficiency, but it may be impossible due to the physical arrangement of the system. Increasing the capillary diameter would increase efficiency and reduce transit times, but a larger skimmer pump would be necessary to accommodate the increased gas flow.

Perhaps heating the target chamber or pre-heating the He would help to accomplish both results. The effects of the increased temperature on cluster stability and on the cluster-recoil attachment mechanism are uncertain, but an investigation of this parameter may have rewards.

The connection of the capillary to the target chamber could definitely be improved. The best way may be to insert the teflon capillary directly into the chamber, eliminating the stainless steel tube and the discontinuity at the point where the teflon tube presently slips over the stainless steel capillary.

If further investigation reveals that cluster volatility is still a problem, a new cluster material of lower vapor pressure should be sought. The successful use of NaCl clusters elsewhere [24, 74] indicates that this may be an appropriate substance.

Using a HeJRT system to supply TRISTAN with non-volatile fission products is a novel but meritorious approach. The successful achievement of such a union would signal the beginning of a new era for on-line isotope separators.

Although it is not possible to claim in this thesis that a successful coupling has been made between a HeJRT and mass separator ion source, the steps remaining to accomplish the task seem clear. The ion source should be redesigned to ensure that all internal parts are at a high enough temperature to prevent condensation of non-volatile fission products in the ion source. Then, the various parameters of the coupled system should be optimized to maximize transmission efficiency of the system. Finally, the coupled system should be combined with mass analysis of the extracted ion beam in order to ensure that there is no effect on the performance of a mass separator by use of the HeJRT coupled to the ion source. Unfortunately, it is not possible to predict the ease or difficulty with which these remaining steps might be accomplished. If past experience is to be any guide, surprises are in store during the continuation of this work.

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