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SOME ADVANCES IN NEGATIVE ION TECHNOLOGY

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Introduction

Although knowledge of the existence of negative ions as a possible state of nature is well known from chemical transport theory and also from the parabola observations of J. J. Thompson,[1] negative ions have proven to be rather elusive particles. Consequently, most physicists have tended to consider them laboratory curiosities. The dominent reason for the elusiveness of these particles is that their electron affinity is almost never greater than 3eV so that as entities they are fragile and can be readily dissociated by collisional detachment during interactions with other molecules.

Notwithstanding these problems of formation and storage, there is currently considerable interest in experimental techniques which will allow the production of intense beams of negative particles from elements throughout the whole periodic table. As all of us at this conference know, tandem accelerators have provided for more than a decade the major application of negative ion technology; and later in this session, Dr. Voss will be describing the needs for negative ions of the large new tandem which will be built at Daresbury. Within the last few years, however, these tandem applications have been moving away slightly from the center of the stage and there is growing needs for intense negative beams in other fields. For example, there is interest in alternate phase acceleration for meson factories, for efficient high energy neutral beam injection into fusion reactors and in other areas which are probably outside the scope of this conference. These other areas include the construction of supercollimated beams, high energy ion implantation, surface studies and analysis, and the simulation of neutron induced recoil damage in reactor fuel elements.

During the past two decades, the advances which have been made in the techniques of negative ion production have been truly spectacular both in terms of intensity and the available species. For example, while in 1955 negative ions of the hydrogen isotopes were produced at submicroampere intensities, today 22mA beams of negative hydrogen have been reported[2] and it is rumored that Dimov and his collaborators at Novosibirsk have seen 200 milliampere peak pulse intensity during pulsed H^- operations. Equally spectacular has been the increase in the variety of species which has occurred during the past 20 years. This increase can be seen from Figure 1 which is a periodic table with those atoms that have produced useful (microampere or more) beams of negative ions being heavily outlined.

Much work still remains to be done on improving the performance of negative ion sources and the purpose of this talk today is to describe some of the properties of negative ions, to discuss ways in which the properties of potential negative atoms or molecules may be predicted, and to describe some of the recent advances in source technology. There will be little discussion of the details of classical negative ion sources as these were covered thoroughly by Dawton[3] in the proceedings of the 1971 Heavy Ion Source Conference.

Negative Ion Properties

The subject of negative ions and their properties were reviewed by Massey[4] in 1950. More recently Berry[5] has reviewed the properties of atomic and small molecular ions and an article by Steiner,[6] dealing mainly with photo detachment, provides extensive information about negative ion affinities.

Figure 1 is a periodic table which shows first ionization potentials and electron affinities. The upper number in each box is the ionization potential and the lower the electron affinity. (The electron affinities were compiled from refs. 5, 6, 11). It can be seen that more than 75% of the elements have a positive electron affinity so that it should be possible for them to form stable negative particles in the appropriate environment.

For many accelerator applications, it is not essential that one uses negative atomic particles. It is often equally useful to include molecular compounds as potential negative candidates. For example, in a tandem a negative molecule is almost as effective providing the companion particle is small in mass compared to that of the parent particle. This possibility of compound acceleration extends the possible negative ion species even further so that the 75% availability mentioned in the previous paragraph is made almost 100%. A good example of compound acceleration is nitrogen where one can routinely produce several microampere intensities of NH^-[7]. At first sight the noble gas atoms would appear to be poor candidates for negative particle formation. However, it is interesting to note that ArF^- has been found in mass spectrometry studies[22] and may well indicate a way in which Ar can be accelerated by a tandem. Table 1 gives the electron affinities of a few compounds which might be interesting to accelerator users. It should be emphasized that this list is just a tiny sample of known negative compounds.

Table 1

Ion Species	Affinity[8](eV)
BeH^-	2.85
$BeH^=$	possibly stable
BH^-	0.09
BH_2^-	stable
NH^-	0.22
MgH^-	1.08
ALH^-	0.04
SeH^-	2.0
WO_3^-	3.6
$C_6H_5^-$	2.2
SF_6^-	1.49
$C_6H_5CH_2^-$	0.90

In addition to the stable configurations mentioned previously there is a third class of negative particles where an electron is attached to a neutral particle that is not in its ground state. This class includes the metastable negative ions, the most famous of which is He^- which has a lifetime of 18.2 microseconds.[9] It is believed that during formation of this particle the neutral He atom becomes trapped in an excited triplet state whose decay to the ground state cannot take place without an electron spin flip. During this period, while the neutral helium is excited, a third electron can be attracted to the atom and be bound to form a negative ion. While this procedure sounds complicated and unlikely to be useful, reliable equipment is now available which permits routine formation of 10-20 μA beams of He^-.

Negative Ion Calculations

While the previous remarks indicate that useful negative beams can be developed for almost any element, it is essential for any such development that one has a good idea of the negative species which are likely to be stable. In principle, the electron affinity of atoms in their ground state can be calculated by various Hartree-Fock procedures. However, because of the small size of most electron affinities, extremely accurate calculations must be made to get definite evidence of stability. Such calculations are difficult, time consuming and require major computational facilities. A number of theoretical groups have attacked these challenging problems. Weiss[10] has studied the electron affinity of Li, Mg, K, and has derived an electron affinity for Li of 0.62 eV which is in excellent agreement with the experimental value of 0.6 eV. Clementi[11] and his collaborators have made a number of calculations which are accurate to 15% and can be considered reliable guides for those atoms which can be expected to form negative particles.

Recently, a number of calculations for molecular systems have been made with high precision by Koski, Kaufman and their collaborators at the John Hopkins University. In this quest for new negative species, the John Hopkins group have constructed major computing programs to allow large calculations of the Hartree-Fock-Roothaan type to be carried out with high precision. As an example of the accuracy of these calculations, the Hopkins group have calculated[12] the vertical electron affinity between O_3^- and the neutral O_3 molecule. This is clearly an elaborate and time-consuming calculation because it is not only necessary to relax the system for a particular geometry, but also to vary the bond angle and separation around the equilibrium values. The final calculated value of 2.12 eV is in excellent agreement with the unpublished photo detachment measurements of Beatty[13] which gives an electron affinity of 2.1 to 2.2 eV. The Hopkins group has also made calculations showing that the molecular BeH^- is stable and that the molecular $BeH^=$ also is probably a stable entity.[12]

Calculations are invaluable as a guide for designers of negative ion systems and I believe that every encouragement should be given to theoretical groups by all of us that are interested in negative particles so that the techniques can be extended and negative calculations made for more complicated molecular species.

Negative Ion Catalysis

In the history of the production of useful beams of negative ions, there is a landmark paper by Donnally and Thoeming[21] which deals with the specific problem of He^- production. At the time that this paper was published, He^-, like most other negative species, had been produced previously only by charge exchange collisions between He^+ and various permanent gasses (He, Ne, Ar, Kr, O_2, N_2, and H_2). In such collisions the cross section for production of He^- is small and of the order of 10^{-19} cm^2 leading to an He^- equilibrium fraction of approximately 2×10^{-4}. Because the production is probably a two-stage reaction, the individual cross sections for both neutralization and electron addition must be high. An important point which Donnally made was that if in the electron addition stage the charge exchange atom for helium is chosen such that for large inter-nuclear separations the potential energy curve of $He^- + A^+$ is just a few eV above the potential energy curve for the neutral combination $He^\circ + A^\circ$ then the Coulomb interaction will tend to bring these potential energy curves together as the internuclear separation decreases with the possibility of mixing and crossing present. When this condition is satisfied, the Massey criterion[14] suggests that the cross sections for exchange can be large. This argument implies that a suitable charge exchange molecule should have an ionization energy which is only slightly greater than the electron affinity of the final electron in the negative ion.

If we look now at the periodic table in Figure 1, it can be seen that the alkaline metals cesium, rubidium, etc., satisfy best this criterion. The first ionization potential of cesium is only 3.87 eV and Donnally showed that Cs can be used effectively to produce He^- ions.

There is no reason to limit the argument of the previous paragraph to He^- production and in practice the same type of mechanism can be used to enhance the production of many species of negative particles. Earlier we mentioned the rumor that Dimov had produced 200 milliampere of negative particles at Novosibirsk. It is our understanding that this was achieved from a comparatively standard duoplasmatron arrangement with cesium present in the gas.

Krohn[15] reported in 1962 that the yield of Cu^- ions could be increased by a factor of 10 when cesium was sprayed onto a sputter surface. This fact was exploited in a source system by Hortig[16] whose operation is shown schematically in Figure 2. The source consists of a rotating wheel into which is embedded the material from which negative ions are required and onto which is coated continuously a thin layer of cesium. An Argon ion beam injected at a small angle sputters the source material away so that the low energy positive ions pass through the thin monolayer of cesium on the surface, are charge exchanged, and then accelerated by the extraction geometry.

Figure 3 shows schematically the geometry of a lithium ion source which has been in operation at the University of Rochester for several years. The principle of operation is that a heated boat supplies lithium vapor which coats the inside of the canal which is bombarded by a helium or argon beam. Once again, the lithium ions that are sputtered by the incident particles travel through the surface layers and are charge exchanged to negative and accelerated. Currents in excess of 5 microampere have been produced and the source operates reliably between 2 and 4 microamperes for periods of ten days or more.

At the SNEAP conference[17] in Tallahassee, there was considerable discussion by Middleton and Purser on various forms of surface sputter sources. The upshot of these discussions was that a source similar to that shown in Figure 4 was constructed at Rochester to produce a wide variety of negative ions. The principle of operation is that an argon beam coming from a standard duoplasmatron is used to bombard the inner surface of a sputter cone which is coated with cesium from an alkaline metal evaporator. An electric field produced by the cylindrical electrodes extends into the conical region to produce an electric extraction field which accelerates negative ions that are produced in the source and charge exchanged in the surface layer of the cesium or lithium.

The next important step in this ion source development program was taken by Middleton,[18] who installed a surface ionization source for potassium in place of the argon duoplasmatron reported in the previous paragraph and mounted an extraction cone on a wheel so that it is possible to have up to 12 different species of negative ions available from a given source without breaking vacuum. The principle of this device is shown in Figure 5.

The operation of surface ionization sources has been described by Daley and Perel[19] and by Kuokevics and Thompson[20]. In operation, the tungten frit which makes up the emitter of the surface ionization source is maintained at approximately 1200°C with a partial pressure of potassium or cesium of a few torr maintained behind the cell. Under these conditions, the alkaline metal will diffuse through the surface and will be ionized to a degree which is dependent upon the relative work functions of the surface and of the alkaline metal. In practice, if the work function of the surface exceeds that of the alkaline metal by 0.5 eV, close to complete ionization can be achieved. In practice, these surface ionization devices can produce several mA/cm^2 of ionized cesium and there is every reason to believe that the unit will scale with size. Figure 6 is a schematic diagram showing the geometry of the surface ionization unit taken from the paper of Daley and Perel. This particular source was designed for the production of a lithium beam and uses a wick material for transporting the liquid lithium into the heated area. A spray of oxygen is impinged upon the front surface in order to increase the work function of the surface layers and improve the ionization ability. Figure 7 shows the efficiency of the ionizer as a function of surface temperature. It can be seen even at 12 milliampere cm^3 that the efficiency of ionization is approximately 60%.

Table II is a composite of the experience at Pennsylvania and Rochester. The great virtue of these types of sputter systems can be summarized: Firstly, the material that is to be accelerated is not injected into the arc chamber of an ion source so that there are no problems from cathode poisoning and it is not necessary to have elaborate ovens and heater control to provide vapor control. Secondly, this source eliminates the need to learn to operate a new source for each particular element. The basic design uses a standard duoplasmatron or cesium surface ionizer as the basic source of sputtering particles so that one does not have to have a major program for each new heavy ion that must be produced. Thirdly, the source is inherently bright. The energy spreads are expected to be low and we at Rochester have found that beams from such sources are transmitted through our accelerator with very high efficiency. Finally, using the wheel technique, the beam species can be changed in minutes.

Some Other New Negative Sources

It would be unfair to close this talk without some mention of the other important advances that have been made in this field. In Figure 8 there is the cross section of a source that has been developed at Orsay, called the Trio Plasmatron. This source, which has been developed in Septier's

Table II

Beam	Current (μA)	Laboratory	Comments
D^-	2.5-3.0	Penn	Deuterium absorbed on Titanium Cone
B^-	>1.0	Penn	
BO^-	10	Penn	
C^-	10	Penn	
NH^-	1.5	Roch/Penn	
O^-	10-100	Penn/Roch	
S^-	10	Penn	
Ca^-	1.5	Roch/Penn	acceleration of Ca^{44} is possible
Fe	1.0	Roch	
Ti^-	0.5-1.0	Penn	
Ni^-	3.5	Roch/Penn	
Cu^-	8.0	Penn/Roch	
Au^-	3.0	Roch	
Bi^-	0.2	Roch	

laboratory,[23] divorces the arc chamber of a conventional duoplasmatron from the region where the gas is inserted by allowing the plasma to drift from the source and ionize the gas in an expansion chamber which is filled with an appropriate gas mixture. This ion source has already produced 250 microampere of O^- beam and it seems likely that there may be substantially more available.

An interesting modification of the direct extraction duoplasmatron source has been described by Colubev[24] from Leningrad. In the past, beams of negative hydrogen have been obtained by offsetting the intermediate electrode of a duoplasmatron from the extraction aperture. The intensity limitation of this technique is that the primary arc in the ion source tends to erode the anode structure away so that there is a limitation in the maximum beam that can be extracted. Figure 9 shows a hollow tubular anode structure described by Golubev where it is possible to avoid the above disadvantages by producing an arc in an annular region and extracting the negative ions on axis. The source was operated at Leningrad in a pulse mode with arc discharge currents in the range 20 to 100 ampere. In pulse mode, focussed beams of H^- with peak currents of 6 milliampere and low emmitance have been obtained with electron loading currents of less than half an ampere.

Summary

The state of negative ion technology at this time is undergoing a substantial transition. Beams have been produced from a large number of species of the periodic table and ways can be seen which will allow almost any species to be accelerated successfully. Because of the great usefulness of negative ions, I believe that a National effort should be made to exploit the possibilities of these sources by developing the calculational techniques which we described earlier, and by the development of various types of negative ion sources.

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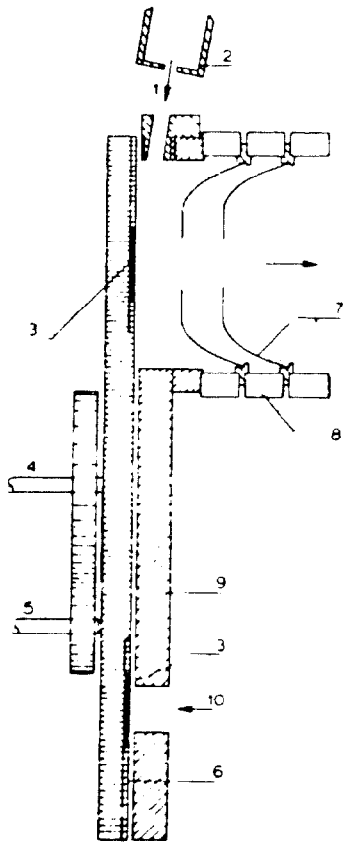


Figure 2.

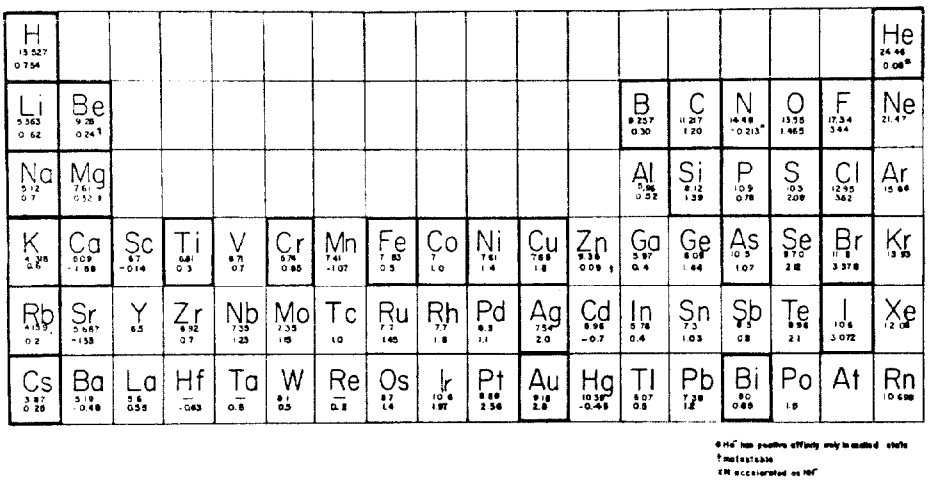


Figure 1.

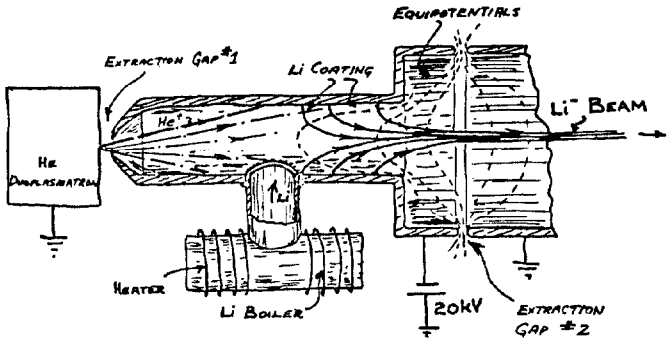


Figure 3.

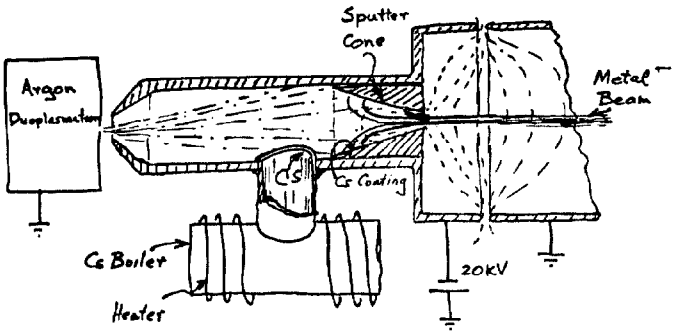


Figure 4.

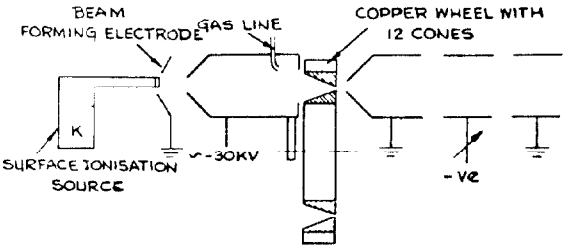


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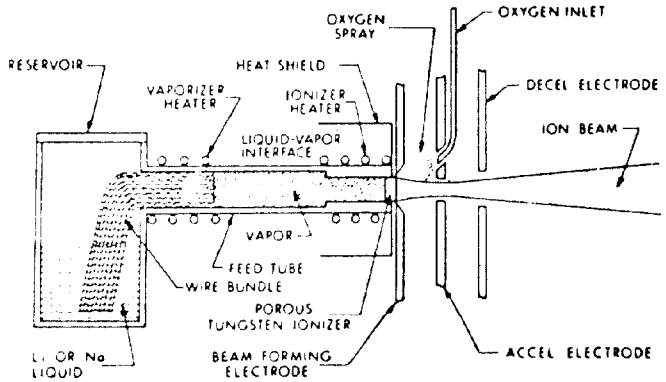


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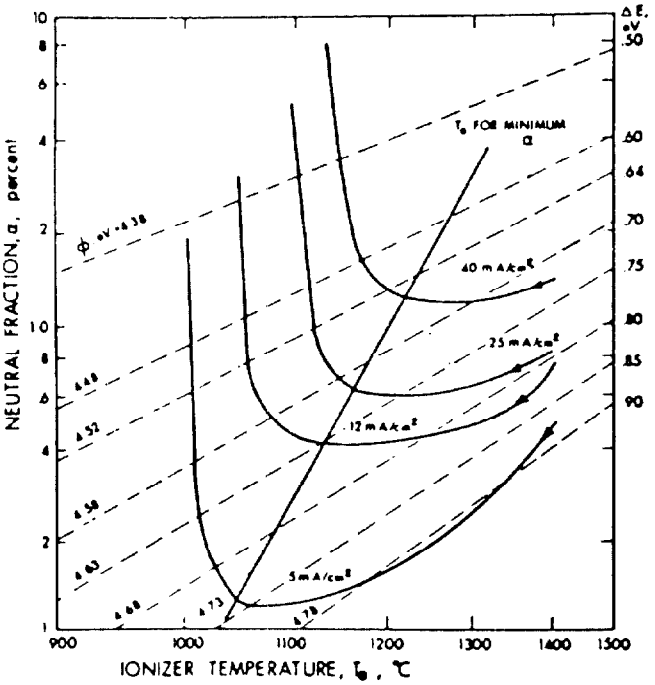


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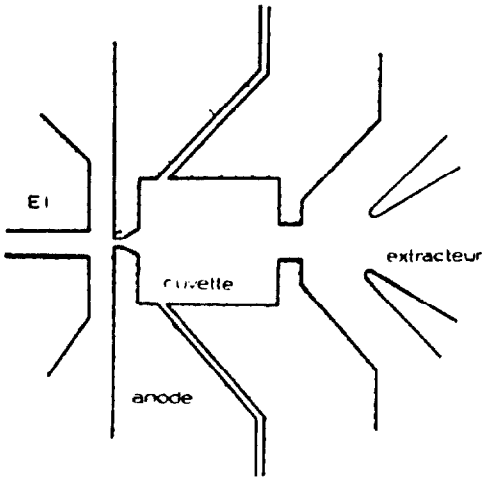


Figure 8.

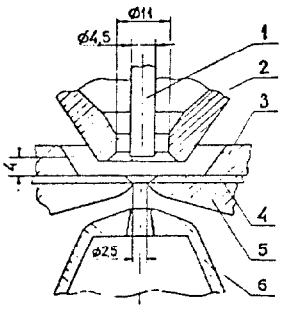


Figure 9.