Application trends for electrostatic ion beam accelerators

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From the 1930’s to the 1960’s electrostatic accelerators were used primarily for nuclear structure research. This has changed dramatically in the decades that followed. This talk will discuss the applications and their effect on accelerator design and performance. The most recent use for electrostatic accelerators is in the field of pharmacokinetics, which is discussed with a tentative look to the future.

Keywords: Pelletron; electrostatic accelerators; PIXE; RBS; pharmacokinetics.

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1. Introduction

As electrostatic accelerators moved away from the single application of nuclear structure research, new applications required additional capabilities not present in the dedicated accelerator used for nuclear structure.

General advances in overall technology led to significant improvement in the electrostatic accelerator itself from the original design of the modern accelerator column structure [1]. These included such things as the move away from belt charging to the insulated Pelletron charging chain and the multimillion volt power supply. Stepper motors with nylon line for control of components in the terminal gave way to light links with programable power supplies. This led to the development of modern double shielding techniques. In addition, better computer systems allowed better ion beam optics calculations and assisted in the design of better ion sources, beam line optics and acceleration tubes.

Outside of the accelerator pressure vessel, new applications are dictating specific beamline configurations, detector systems, and accelerator control systems combined with data collection software. These applications have moved well beyond the nuclear research field into areas as diverse as archeology, climate studies, pharmaceutical research, and many others.

2. From the 1930’s into the 1970’s

The electrostatic accelerator progressed from a metal dome on a tripod-like structure to the modern column structure presently in use developed in the 1930’s [2]. The first comprehensive scattering experiment, protons on protons, was published in 1939 by Herb, Kerst, Parkinson and Plain at the University of Wisconsin [3]. At this time, there was no commercial supplier of electrostatic accelerators and the only multimillion volt systems were single ended accelerators. This changed in the late 1940’s.

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3. From the 1970’s into the 1980’s

In the 1960’s Dr. Peter Rose, Chief Scientist at HVEC, and others started to investigate the use of heavy ion keV beams for materials modification to change the conductivity in silicon [5]. This was the beginning of production ion implantation and an early major step in decreasing device sizes in integrated circuits [6]. As with nuclear structure research, the beam energy requirements continued to increase until, in the early 1980’s, production implanters were required with ion beam energies in the 2 to 4 MeV region. In addition, there was a move away from 100 s of nanoamps of beam current into the milliamp range of beam current in order to provide adequate throughput for silicon wafers.

While it was typical of the nuclear structure research to have a 3 or 4 day run after which the ion source was com-
pletely rebuilt, this method of running would not be suitable in a production environment. There was a very strong emphasis on total up time and best possible reliability. For the first time, electrostatic accelerators would be operated by individuals with very little knowledge of the operations of the accelerator itself. Therefore, the design of the complete accelerator system had to address these issues of uptime, reliability, and ease of operation while maintaining rapid throughput of an implanted product.

From 1981 through 1985, 8 systems based on a 1 MV tandem Pelletron were sold to process silicon wafers with diameters from 4" to 6". The systems were completely enclosed and great care had to be taken in the design of the accelerator to limit radiation. For the first time, electrostatic accelerators were not housed in a radiation shielded vault but on a factory floor. An ion beam rastering system had to be designed to provide a uniform dose across the entire silicon wafer surface. This led to the inclusion of a neutral beam trap to prevent neutrals from reaching the wafer surface.

Today, the market for production MeV ion implanters is still strong. They are based on LINAC systems as well as solid state power supply based tandem accelerators. However, the accelerator itself is a very small part of the overall system. The main concerns are wafer handling, heat management during the implant process, particulate control and monitoring, elimination of surface charging during implantation, as well as the continuing concern for maximum uptime and throughput.

4. **From the 1980’s to the present**

While ion implantation was being investigated in the 60’s and 70’s, techniques developed for nuclear structure research were beginning to be applied toward materials analysis [7]. Techniques which were very simple for the nuclear physicist such as elastic scattering (Rutherford Backscatter), elastic recoil detection, and nuclear reaction analysis were now being considered for elemental analysis for a wide variety of materials analysis applications [8]. As with ion implantation, this meant moving the electrostatic accelerator out of nuclear physics into other fields where accelerator operation was not well known. The development of commercial RBS, ERD, NWA and PIXE spectrometers began in the early 1980’s. There was a very strong need to be able to analyze semiconductor materials for quality assurance and applied research. Again, as with ion implantation, there was increased pressure for rapid throughput of samples being analyzed. This lead to the development of analysis end stations which could accommodate channeling and a variety of MeV ion beam analysis techniques. In addition to the hardware, software had to be developed both for data collection and accelerator control. It became desirable to set up the accelerator system to analyze samples while the system was completely unattended.

In addition, a reliable ion source for the production of the He\(^{-}\) ion beam was required [9]. This lead to the development of the RF charge exchange ion source which does not have the disadvantage of a hot filament as in the older duoplasmatron charge exchange system. There are now almost 100 of these ion sources in operation.

Both RBS and PIXE have evolved into greater capability with the use of microprobe lenses and high resolution detector systems. Microprobe lenses for both of these applications are now commercially available to allow beam spot sizes from the tens of microns to submicron range. This allows the elemental mapping of very small structures including biological cells. Micro PIXE and micro RBS require a beamline that has a very long object distance for the microprobe lens, 4 to 8 meters, and a very short image distance. The overall result is a microprobe beamline length of up to 8 meters.

The demands of the submicron beam for micro PIXE require a very good ion beam optics design, a high brightness source and low beam energy spread. While both tandem and single ended accelerators are used, often it is the single ended accelerator that is preferred. A tandem accelerator will have an additional energy spread contribution from the negative ion to positive ion stripping process.

The property of a low beam energy spread is also required for the high resolution RBS Fig. 1. shows a high resolution detector system using a 90 degree magnet to analyze the scattered secondary ions. These high resolution systems are designed to look at the first few atomic layers. The detector resolution is typically on the order of 1 keV or less. In order to prevent an ion energy spread of larger than this, the beam penetration into the sample must be limited. Therefore, beam energies below 500 keV are typically used for high resolution RBS applications. The result is angstrom level resolution.

Analysis techniques based on MeV ion beams are now routine. They offer a sensitivity not previously available using low energy ion beams and other techniques [10]. Based on the number of analysis end stations that have been sold, the number of accelerators from 1 MV to 3 MV in use, and the number of He\(^{-}\) ion sources also in use, it is estimated that the number of active RBS/PIXE laboratories is near 100.

5. **From the 1990’s into the present**

The most complex demand on the design of the entire accelerator system came in the 1990’s with the new popularity of Accelerator Mass Spectrometry (AMS) systems. Although AMS was begun in the early 1980’s and 5 dedicated systems were sold, it did not gain wide spread acceptance until the 1990’s when backgrounds and precisions were greatly improved. The first application for AMS was carbon dating.

Before the use of AMS, carbon dating was done by decay counting of \(^{14}\)C which has a half life of about 5,730 years. The disadvantage of decay counting is that you must wait for the decay to occur. For modern carbon, there are about 13.5 disintegrations per minute per gram. Therefore, counting for a 1 gm sample to about 1% statistical precision of \(^{14}\)C takes about 12 hours. In contrast, modern carbon gives 9,000
to 11,000 $^{14}$C events per minute for a milligram sample at the detector of an AMS system. The end result is that AMS requires a much smaller sample, a milligram to 100s of micrograms, while providing a much faster time to the required statistical precision.

For most cases, AMS is the measurement of the ratio of a radioisotope to an abundant isotope [11]. Therefore, in the case of carbon, $^{12}$C, $^{13}$C and $^{14}$C are all measured to high precision. This puts extreme demands on the complete design of the overall accelerator system. In the high voltage terminal, gas stripping is recommended. A carbon foil will change thickness over the time, changing the beam energy. The gas stripper target must be thick enough to allow for equilibrium stripping so that small changes in gas pressure do not affect the charge state fraction and all hydrocarbon molecules are broken up. For compact AMS systems, approximately a 2 $\mu$g/cm$^2$ gas target is needed. This calls for an extended gas stripper tube with two turbo molecular pumps in the high voltage terminal. The ion source must be capable of handling a large number of samples. This ion source is a cesium sputter source, which accommodates 40 to 200 samples in a vacuum volume at one time depending upon the ion source type. The injector must be designed with a 90 degree magnet with a true image and object position of the highest possible resolution. Typically, sequential injection is used by biasing the injection magnet chamber. It is recommended that all masses of interest be measured. For carbon, this includes mass 12, mass 13 and mass 14. At the high energy end, another 90 degree magnet is needed with a wide exit pole to allow the collection of the abundant isotopes while allowing the rare isotope to proceed for further electrostatic analysis and on to the detector system Fig. 2.

As mentioned above, AMS started primarily for carbon dating. AMS is now used for a wide variety of radioisotope measurements including those of beryllium, aluminum, calcium, chlorine, iodine and many others including the actinides. All of these measurements require a significant amount of sample preparation. The sample must be suitable for use in the cesium sputter source. In the case of carbon, graphite has been the primary material. However, some labs are now using CO$_2$ directly to avoid the complication of producing graphite [12].

Today, there are about 70 AMS laboratories worldwide. While carbon dating and related applications are still dominant, at least 8 of these laboratories are dedicated to biomedical applications.
6. The present and beyond

Electrostatic accelerators have been just behind the headlines in many high profile applications for many years. Table I. gives a list of some of these examples. It is expected that this will continue to be the case for many years to come. One example is in the field of pharmaceutical research, drug discovery. Specifically, the field of pharmacokinetics relies heavily on AMS techniques.

Pharmacokinetics is the study of the drug/organism interaction involving the investigation of ADME processes [13]. By measuring the level of a radioisotope in a radiolabeled biological sample, information can be obtained concerning the bioavailability of a drug (A), the distribution of the drug in the body (D), the metabolites formed by the body from the parent drug (M), and the elimination rate of the drug from the body (E). This work was pioneered by Vogel et al. at the Lawrence Livermore National Laboratory [14].

In the past, radiolabeling used decay counting as in carbon dating. In order for decay counting to be effective in a reasonable time, a large amount of the radioisotope had to be added resulting in a significant amount of radioactive waste. However, the extreme sensitivity of AMS allows a very small amount of the radioisotope to be added so that the increase in radiation is very near ambient. At the same time, results for a given precision can be obtained in a matter of minutes compared to a matter of weeks in the case of decay counting [15]. One very significant result is that now radiolabeled drugs can be used in human volunteers. This has lead to a number of advancements by allowing a direct comparison between animal models and human studies. It has also decreased the time to bring a drug candidate to market by allowing first in human studies far earlier [16].

In one area, metabolite profiling, AMS allows the detection of metabolites that could not be seen by any other method. This is very important in understanding the effect of a given drug on the body including side effects. Typically, a blood sample is analyzed by taking a blood sample from a human volunteer who has ingested a radiolabeled drug. The sample is passed through a Liquid Chromatograph (LC) and the resultant mass fractions are each converted to graphite and analyzed via AMS. However, each profile can have as many as 100 to 200 mass fractions. This results in a very time consuming and expensive sample preparation for each metabolite profile.

This application and others have lead to the development of using CO₂ instead of graphite as mentioned above. The mass spectrometry laboratory in the MIT department of Biological Engineering under the direction of Professor Steve Tannenbaum [17], has developed a method of direct, inline combustion of liquid samples from the LC for injection into an AMS ion source. They have used their laboratory system to investigate total ¹⁴C in individual mass fractions. National Electrostatics Corp. is now licensed to commercialize this system and make it available for use on modern AMS systems.

While the use of AMS in drug research has increased the complexity of the accelerator system and led to the combining of the accelerator with other analysis techniques from other fields, such as the LC from chemical analysis, it has also led to a significant simplification. Modern AMS systems are based on the tandem configuration. One purpose of the tandem is to break up the negative molecules coming from the AMS ion source. The high energy tube of the tandem is required in order to bring the ion beam down to ground potential for further analysis. However, the high energy tube can be eliminated if the post stripping analysis and detection systems remain at the terminal potential. This has been done with the Single Stage Carbon AMS system Fig. 3. [18] There are now 8 of these systems in operation. They have demonstrated the same precision as the carbon compact tandem based systems for carbon AMS.

7. Conclusion

In the more than 70 years since the first air pressurization of a Van de Graaff generator, electrostatic accelerators have provided essential and unique contributions to the field of nuclear structure. The techniques developed in this field with accelerators have made and continued to make unique contributions to essentially every field of modern science. The demands placed on the electrostatic accelerator system have led to significant improvements in ease of use, reliability and overall capability.

Electrostatic accelerators have a unique property of providing multi MeV ion beams through a direct potential drop. The result is a true DC beam with a low energy spread with a much higher degree of control over the properties of the ion beam spot at the target than by any other acceleration process. This allows the development of nanometer diameter beams, systems with extremely low background as in the case of AMS above and the addition of pulsing and bunching for precise nanosecond pulsed ion beams. Therefore, future
applications that require these properties will also require the electrostatic accelerator.

Based on the expansion of applications over the last 30 years and the advances in electrostatic accelerator technology, it is not possible to predict what the new applications or accelerator configurations will be. However, it is highly likely that the electrostatic accelerator will continue to be used in new and unexpected ways.