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INDUSTRIAL APPLICATIONS OF SMALL ACCELERATORS

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Summary

Current practical applications of small accelerators are summarized. These include: trace element studies with both neutron activation analysis and charged particle induced x-rays, accelerator produced neutron well logging techniques, and accelerator-based research associated with the controlled thermonuclear research program. A rather extensive bibliography is included in all three areas.

Introduction

In the United States, there are approximately 500 positive ion accelerators with terminal energies less than 3 MeV. In addition to these, there are approximately 950 electron machines in this same energy range. Most of the accelerator facilities mentioned in this census are devoted to practical applications in medicine and industry. In the last several years, some of the university facilities have started programs in which practical applications, rather than basic research, seems to be the main thrust. This move has been prompted primarily because of the fact that federal funding for basic research with machines in this energy range has been drastically reduced. In fact, AEC (ERDA) and NSF, who have classically supported these facilities, have withdrawn facility support in most cases for basic nuclear research. A wide variety of practical applications with accelerators are being studied in industry, government laboratories, and universities throughout the United States. In this paper, we will point out three specific areas in which there seems to be an abundance of activity. These are: trace element studies, neutron oil well logging, and controlled thermonuclear research. A brief summary will also be given of several other areas that have recently received a great deal of attention. The data contained in this summary has been extracted mainly from a recent conference (October 1974) which was sponsored by ERDA, NSF, NBS, and North Texas State University. The conference was titled The Third Conference on the Use of Small Accelerators in Research, Teaching, and Industrial Applications, edited by the authors of this paper. Copies of the two volume proceedings can be obtained by writing the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U.S. Department of Commerce, Springfield, VA 22151.

It is interesting to note that most of the accelerators in the world are small (less than 3 MeV) according to our definition. Figure 1 taken from Ref. 1 shows the world population of accelerators in 1972.

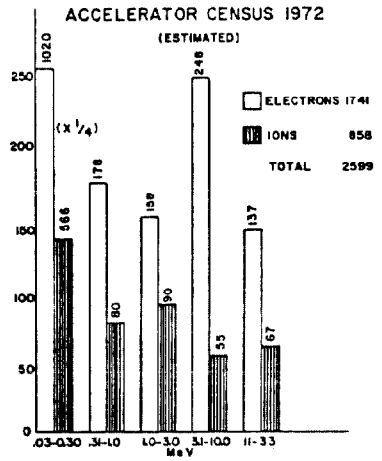

Fig. 1. Accelerator Census

Most of the accelerators that have been sold since 1972 have gone into industry. Most of these are electron beam welding devices and the rapid expansion of machines into the medical profession for radiation therapy. A number of new machines have also recently gone into oil exploration, industrial ion implantation, and radiation processing.

As was mentioned above, this paper will summarize some of the active areas in the technological use of "small accelerators."

Trace Element Studies with Small Accelerators

Part I-Neutron Activation Analysis with Small Accelerators

Nuclear techniques have been used for years in the analysis of trace quantities of elements which are

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present in samples. Neutron activation analysis using primarily 14 MeV neutrons from the $T(d,n)^4He$ reaction or thermal neutrons from a reactor has turned out to be, for some trace elements, the most powerful analytic technique at our disposal. Most of the accelerator work with this technique has been done with Cockcroft-Walton accelerators and energies below 300 keV. For the interested party, many summaries of the theoretical and experimental considerations in neutron activation analysis have been given[1-11]. A review of the literature by S. S. Nargolwalla[13,14] indicates that fifty or so elements have successfully been determined in the concentration range from 10 ppm to 50% by weight. Most of the industrial interest, however, has been concentrated on Oxygen (∿33%), Silicon (∿14%), Chlorine, Calcium and Copper (all about 6%) and other elements such as Barium, Praseodymium, Uranium, and Plutonium. Oxygen and Silicon, of course, are of primary interest to the steel industry. Figure 2 (taken from Ref. 13) shows the distribution of publications in which neutron activation analysis is used. There are approximately 575 accelerators (positive ion) in the world with energies between .03 and .3 MeV.

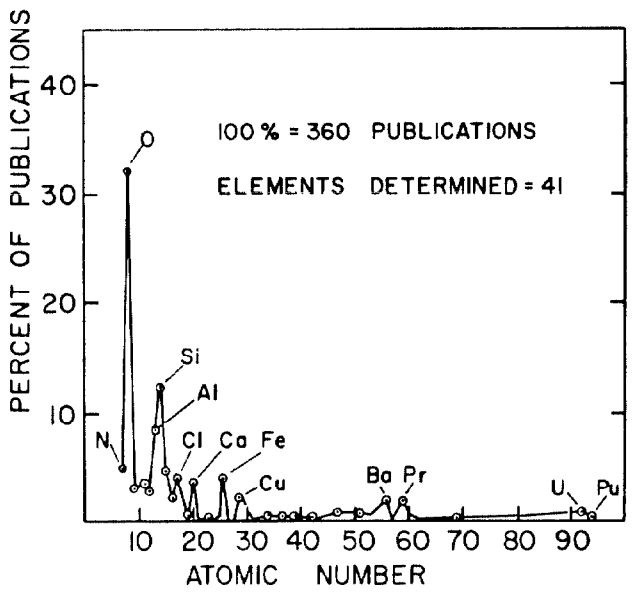


Figure 2

Frequency of Analysis by Accelerator Produced Neutron Activation Versus Atomic Number (Taken from Ref. 13)

It is estimated that there are approximately 200 installations in the world that have neutron activation programs with these small accelerators. The oxygen analysis (mainly in the steel industry) mentioned above is accomplished via the $^{16}O(n,p)^{16}N$ reaction. ^{16}N beta decays to levels in ^{16}O, which in turn decays by the emission of 6.131 and 7.115 MeV gammas. By counting with a NaI detector all events above 4.5 MeV, the investigator is fairly certain that he is counting oxygen gammas. There are a few elements found in steel that cause interference, but this is not usually the case. This non-destructive process for oxygen analysis is inexpensive, rapid and capable of on line use which is very important from the industrial point of view. Several papers have been written which are addressed to specific industrial problems associated with on-line applications[15-18].

Many scientists predict that there will be an expanded use of accelerator activation analysis in the areas of environmental and bio-medical studies. This method is ideally suited for the analysis of many trace elements which exist in organic matrices. Oxygen, Nitrogen, Sulfur, and Phosphorus have been studied in biological samples and represent those elements which can be done with ease for these samples[19-21].

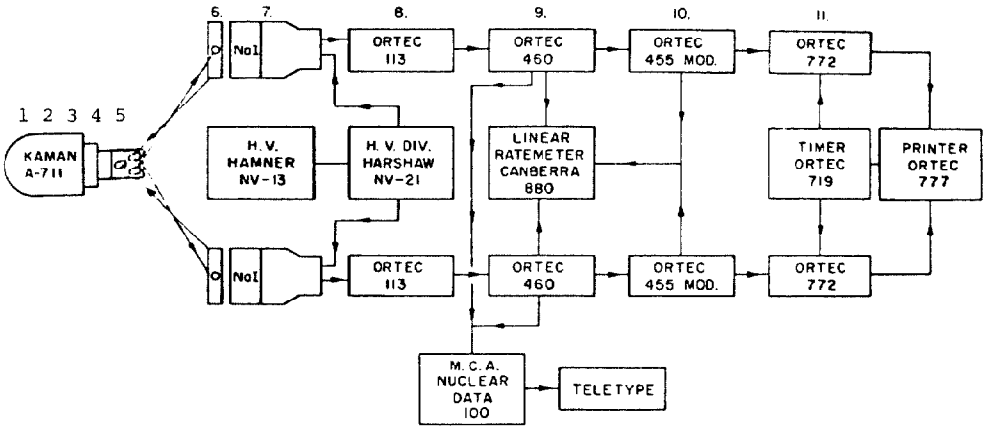
There are many applications in which sensitivity is not as important as the absolute magnitude of the element that is being studied. Examples of this are given by W. D. Ehman et al.[22] for major elements in lunar material. Precision experiments by F. Burns and H. Priest[20,21] on the analysis of silicon nitrides (Si_3N_4) and (Si_3N_4) have also shown that neutron activation analysis is perhaps the most precise way of doing oxygen content analysis of these lighweight materials. Figure 3 shows a Dual Activation System that is currently being used by A. Volborth et al.[23] at the University of California, Irvine. The dual rotating target system removes bias by making corrections for radioactive decay unnecessary. This is accomplished by activating a standard and the unknown simultaneously. This method has been used for oxygen by this group with accuracies of less than 1%. Once the system is set up, the analysis is quite simple.

The other trace element technique that is now being used in many accelerator laboratories is proton or alpha particle induced x-ray fluorescence, or more properly, charged particle induced x-ray emission. This technique will be discussed in the next section of this paper.

Part II-Charged Particle Induced X-Ray Emission as an Analytic Technique

In the last several years, considerable interest has been developed in the use of heavy ions to produce characteristic x-rays for elemental analysis. In a sense, this method is similar to x-ray fluorescence (XRF). XRF has been used as an an analytic method for the detection of trace impurities for the last twenty-five years. Basically, XRF involves bombarding a sample with photons while observing the characteristic x-rays from the sample. For XRF, the photons are usually produced either from a radioactive source or an x-ray machine. The recent development of the "Lithium Drifted Silicon [Si(Li)] detector" has completely revolutionized the old art of XRF. With this method and Si(Li) detectors, it is now possible to do simultaneously, qualitatively, and quantitatively trace analysis on as many as 20 elements in a sample. This is possible because these new detectors can be made with thin windows or windowless and have resolutions as low as 150 eV for 5 keV x-rays. The method now has wide applicability in biology, physics, geology, industrial problems, and the whole field of pollution analysis. At this point in time, XRF with tube excitation has detection limits which seem to be fairly well established at ∿0.1 ppm.

The main difference in "charged particle induced x-ray studies" and XRF is the method of excitation. We will see that sample preparation and analysis in both methods have many common techniques. The person who is interested in trace studies with ion beams is therefore urged to read the abundance of fine papers on photon induced XRF[24-48].



1. SEALED-TUBE NEUTRON GENERATOR.
2. TARGET.
3. SINGLE-TUBE IRRADIATION PORT.
4. DUAL-TUBE SMALL SAMPLE IRRADIATION PORT AND ROTATION.
5. PATH OF "RABBITS".
6. SAMPLE COUNTING PORTS.
7. 4" NaI CRYSTALS AND PHOTOMULTIPLIER TUBES.
8. PREAMPLIFIERS.
9. AMPLIFIERS.
10. SINGLE-CHANNEL TIMING ANALYZERS.
11. PRINTING SCALERS.

Reactor Facility, Chemistry Department, University of California, Irvine.

Figure 3

Dual Activation Analysis System
Used at the University of California, Irvine
(Courtesy of A. Volborth et al.[23])

The advantage that positively charged particles have over electron bombardment for excitation is that the Bremsstrahlung background is about an order of magnitude less for charged particles. Most of the trace element studies thus far have been done with protons and alphas with energies less than 5 MeV. Under these conditions, the Bremsstrahlung continuum is a distribution which extends out to about 10 keV. It is possible to attenuate the Bremsstrahlung with a thin mylar foil (from 0.5 to 10 x 10^{-3} in) and still count the characteristic x-rays of interest in the experiment. Of course, the mylar causes a filter type cut off in the efficiency curve for the detector. Several papers in the literature[49,50] have been addressed to the subject of efficiency measurements for Si(Li) detectors.

X-ray production cross sections for protons and alphas are seen to rise rather rapidly with energy. A typical yeild curve for protons is shown in Figure 4 for Sc, Cr, Zn, Se, and Sb. These data are taken from Ref. 51 and are a result of thin target measurements. The BEA and PWBA notations on the curve represent various theoretical attempts to describe the interactions[52-55]. Figure 5 shows the corresponding yield curves for alphas[51]. Figure 6 shows ionization cross section versus atomic number and the corresponding theoretical fits for alpha excitation.

In terms of sensitivities for thin samples (Figure 4, for example), elements from Zn through Scandium all have ionization cross sections greater than 300 barns for 5 MeV protons. If one now converts this to the problem of identifying these elements in an unknown thin pollution sample, it can quickly be seen that nanogram quantities of these elements can be seen with what turns out to be reasonable operating

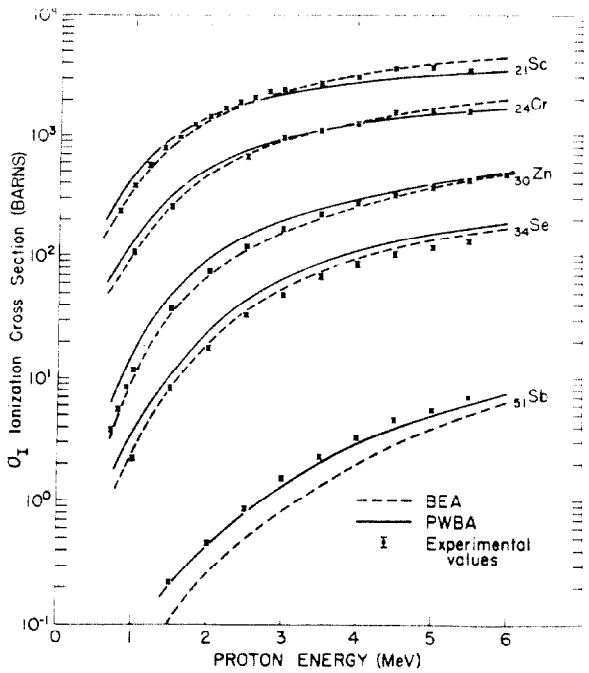


Figure 4. K Shell Ionization Cross Sections for Scandium, Chromium, Zinc, Selenium, and Antimony by Proton Impact

parameters.

Figure 7 shows a schematic diagram for ion beam trace element studies being used by Walter et al. at Duke University. Further details associated with chambers, targets, and experimental procedure can be found by reading the excellent papers which have been produced by the staff members of the dozen or so accelerator laboratories[57-84].

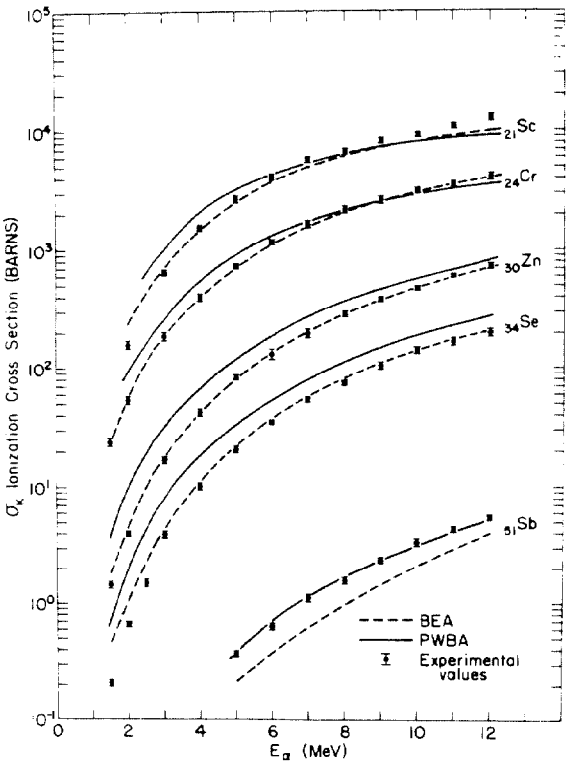


Figure 5. K Shell Ionization Cross Sections for Scandium, Chromium, Zinc, Selenium, and Antimony by Alpha Impact

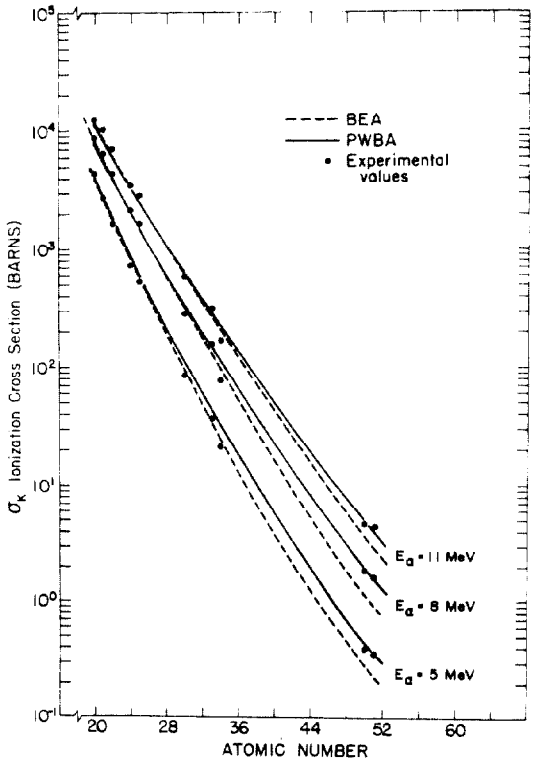


Fig. 6. Alpha Particle Induced K X-Ray Ionization Cross Sections vs Atomic Number for E_α = 5,8,11 MeV

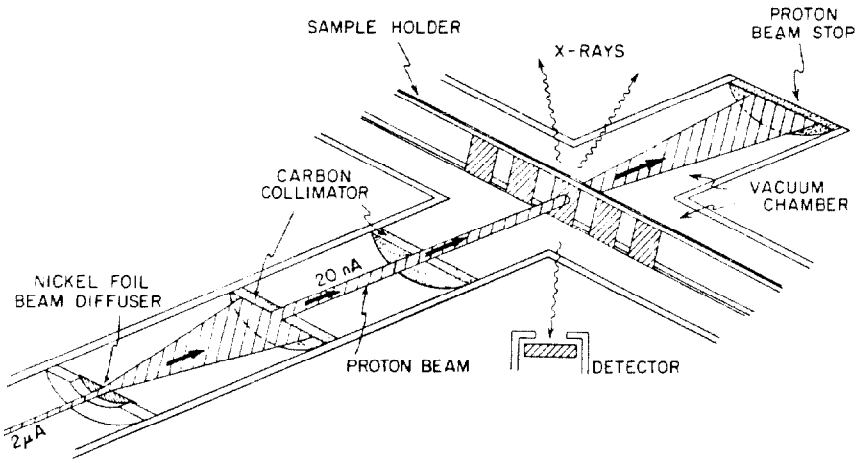


Figure 7

Schematic Arrangement of the Duke University Charged Particle Induced Trace Element Studies - Target Chamber (Taken from Ref. 56)

Figure 8 shows a typical result of some of the work of H. A. Van Rinsvelt et al.[59] from the University of Florida. If one couples together beam current, efficiencies, cross sections, etc., a composite theoretical estimate of the "limit of minimum detectability" can be obtained. Figure 9 which is taken from B. K. Barnes et al., Lowell Tech[64] shows such a plot for thin targets, 3.5 MeV alphas and 3 MeV protons. In terms of practical measurements, sensitivities seem to run from 0.1 ppm for Calcium up to 1.0 ppm for elements as heavy as Strontium. These measured sensitivities are quite adequate for many types of samples. For example, soil samples usually have concentrations of from 1 to 200 ppm of the elements Cl, K, C, V, Cr, Mn, Fe, N , Cu, An, Ga, Br, Rb, Sr, Zr, and Pb. It is interesting also to note that eleven of these elements are found in excess of 10 ppm in most soil samples. The same kind of concentrations are found in most biological samples with usually much higher values for Ca, Fe, and Zn.

The most dedicated accelerator in the United States for charged particle induced trace element

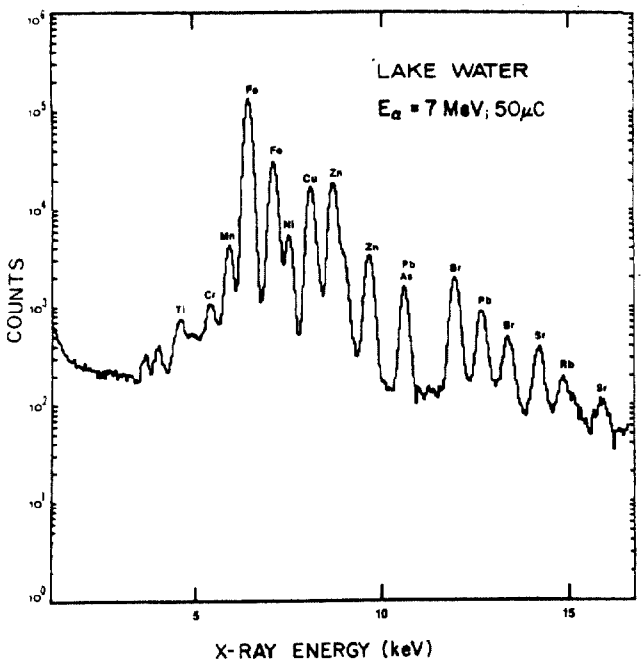


Figure 8

Alpha Particle Induced X-Ray Emission
For Trace Studies of Lake Water
(Provided by H.A. Van Rinsvelt[59])

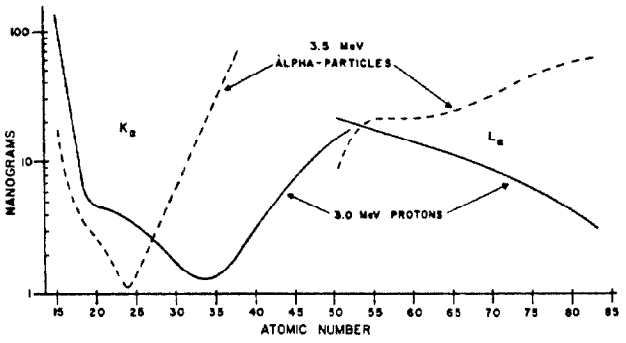


Figure 9

Calculated Limits of Minimum Detectability
For the Lowell Tech X-Ray Analysis System
(Supplied by B.K. Barnes et al.[64])

analysis seems to be the Cyclotron Laboratory of the University of California, Davis. Table I provided by Thomas A Cahill[60] shows typical operating parameters for their aerosol analysis system. In Table I, it should be noted that "ion backscattering" is used for the analysis of elements Hydrogen through Fluorine. This technique is described in Ref. 65.

In summary, the technique of ion beam x-ray analysis coupled with ion backscattering can be used to analyze samples for all elements. The samples which have been studied to date include biological, biomedical, geological, chemical, and environmental specimens. Sensitivities for practical samples seem to lie in the 0.1 to 1.0 ppm range for most elements. The technique certainly rivals most other analytic methods insofar as cost, speed, number of elements for an analysis, and accuracy is concerned.

Neutron Accelerator Well Logging

In these days of the so-called "energy crisis" there are renewed activities in the oil fields. These activities are associated with finding new oil and in determining if there are still usable quantities of oil left in wells that have been shut down because of what was called low productivity before the "energy crisis." In both cases, accelerator well logging is playing a key role in helping to reduce the critical oil shortage.

For most of us, it is hard to imagine the physical constraints that are placed on an accelerator that is lowered into a borehole for the logging process. For this process, Van de Graaffs have been constructed with an outside diameter as low as 3 5/8" and Cockcroft-Walton machines have been produced with 1-11/16 OD. These machines have to be constructed in such a manner that they will withstand pressures up to 20,000 psi, temperatures as high as 400°F and operate at the end of cables that can be as long as 30,000 ft. Most of the logging is done with 14 MeV neutrons from the T(d,n)^{4}He reaction, since it is the most prolific source of fast neutrons for accelerator voltages which one can obtain in the borehole. D.W. Hilchie[84] has described in some detail the parameters of these machines and the method in which the various types of logs are obtained. This method was first used in 1959[85] and has been continually developed since that time[85-96].

There are three basic methods of neutron logging. These are: (1) neutron lifetime logging, (2) neutron activation techniques, and (3) the most recently developed method (1973) of carbon-oxygen logging.

For neutron lifetime logging, the accelerator operates in the pulsed mode. Thirty microsecond bursts[84] are normally produced every one thousand microseconds. After the bursts, the neutrons die away at the rate of two orders of magnitudes in 1000 μsec. Figure 10 shows a graph taken from Ref. 84 of this phenomena. The ratios of the counts under gates 1 and 2 in Figure 10 can be used to determine if oil and gas are present and how much.

Logs based on neutron activation analysis are sometimes quite useful in determining silicon and aluminum concentrations in the borehole and thus give an index of the type of formation, i.e. sandstone, limestone, dolomites, etc. The neutrons from the accelerator

TABLE I

The University of California, Davis Aerosol Analysis System
(Provided by T. A. Cahill, Ref. 60)

1.	Beam Energy and Particle	18 MeV Alpha Beam
2.	Beam Current (Typical)	150 nA
3.	Irradiation Area	0.8 cm^2
4.	Target Backing Parameters	a) 600 $\mu g/cm^2$ mylar b) 1000 $\mu g/cm^2$ Nuclepore
5.	Beam Uniformity	Magnetic sweeping at 1 hz
6.	Irradiation Time	∿100 sec
7.	On-demand Pulsing	200 nsec to off; 50 μsec off 1 pulse
8.	Detectors	a) Si(Li) 10 mm^2; at 90° dΩ = 7×10^{-4} ster Active filter to 1 mm^2 for elements Cl and Below b) Surface Barrier Alpha Detector at 52°.
9.	Live Time	Typically, 75-80% at 5,000 counts/sec, with beam pulsing active.
10.	System Accuracy	±10%, absolute
11.	Minimum Detectable Limits	H→F ∿2 $\mu g/m^3$, Na→Mo ∿3 to 30 ng/m^3, Tin Region ∿300 ng/m^3, Ba→U ∿10 to 30 ng/m^3

TABLE II

Carbon-Oxygen and Silicon-Calcium Well Log Data
(Taken from Ref. 84)

Casing	C/O	Si/Ca	%	Remarks
7"	1.82	1.28	10%	Carbonate
7"	1.74	1.70	28%	Oil Sand
7"	1.61	1.56	28%	Water Sand
5-1/2"	1.56	1.59	30%	Water Sand
5-1/2"	1.66	1.55	30%	Depleted Oil Sand
5-1/2"	1.76	1.62	30%	Oil Sand
5-1/2"	1.74	1.45	---	Limey Sand
5-1/2"	1.64	1.26	---	Shale

activates ^{28}Si and ^{27}Al via the following reactions: $^{28}Si(n,p)^{28}Al$ which decays by emission of a 4.6 MeV β followed by a 1.78 MeV γ the half life is 2.31 minutes; $^{27}Al(n,p)^{27}Mg$ which decays with 4.7 and 5 MeV β's and gammas of 1.013, 0.842, and 0.171 MeV and a half life of 9.5 min. Oxygen which is always present is activated by the $^{16}O(n,p)^{16}N$ reaction, but its half life is 7.35 sec, and the measurements for silicon are made approximately one minute after activation and hence the oxygen activity has died away. If one wishes to study the aluminum a wait period of 15 minutes is required to allow the silicon induced activity to die away.

In carbon-oxygen logs, prompt gammas from excited states are observed. The measurements are recorded for 10 minute counting periods. The neutron source is typically operated in the pulsed mode with 10 microsecond bursts every 10^{-4} seconds. The prompt gammas are observed with a NaI(Tl) detector. In carbon, for example, the 4.43 MeV gammas from the first excited state of ^{12}C and the escape peaks would be observed. Carbon-oxygen and silicon-calcium ratios along with other known data can be used to give valuable information in regard to the borehole. Table II shows part of an evaluation of several measurements using these ratios.

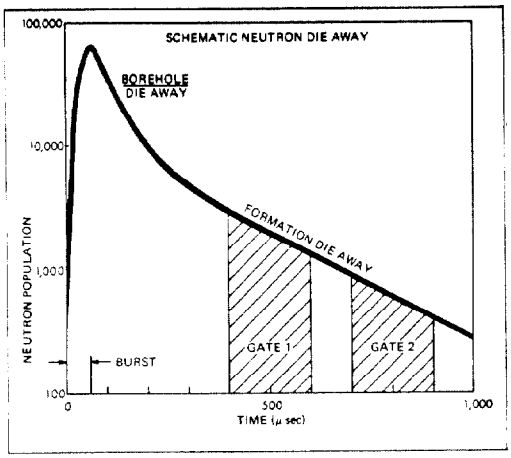


Figure 10

Typical Data For An Oil Well Borehole Neutron Die Away Log[84]

Controlled Thermonuclear Research with Accelerators

It is predicted that between the years 1970 and 2070, the projected world energy consumption will be 5×10^{13} BTU. ERDA is committed to the development of a Controlled Thermonuclear Reactor (CTR) to help supply this gigantic energy demand. Accelerators are playing an important role, not only in the development of the CTR program, but also in the analysis of radiation damage for conventional fusion and breeder reactors. Not only can accelerators be used to study the heating and fueling of plasma for CTR, but they are also being used to study plasma diagnostics, atomic processes involved in plasma buildup and decay, and possible alternate and better nuclear reactions that can be used for thermonuclear power generation once magnetic confinement is proven for the D-T reaction.

It has been pointed out by Barnett of Oak Ridge[97] that since CTR was proposed in the early 1950's, there have been definite needs in the program. Some of these needs along with recommendations from scientists who are knowledgeable in regard to the program have been published in two reports[98,99]. CTR machines as of today have plasma ion temperatures of 300 to 500 eV. Attempts are being made to heat the plasma by injecting neutral hydrogen atoms into the plasma. At present, there is a great need for ion source development to supply these neutral atoms. Beam currents of from 0.1 to several hundred amperes are needed for this application[100]. In order to inject neutrals into a plasma, ions are first made in an ion source, accelerated, then neutralized by passing them into a gas cell in which the neutralization occurs through atomic charge exchange in the neutral gas. Since the charge exchange cross section decreases with increasing ion energy, it is better to keep the initial ion energy low. Then, of course, the energy per atom that is dumped into the plasma is low. This difficulty can be partially overcome by accelerating negative ions into the gas cell at approximately 100 keV and then stripping the electron off by charge exchange. These stripping cross sections are roughly independent of energy; therefore, there seems to be a need for the development of high intensity negative ion sources. A "state-of-the-art" summary of injection ion sources has recently been given by Schilling[100]. Numerous other articles in this regard can also be found in the literature[101-107].

Once the ions are made, accelerated, neutralized and sent into the plasma, they are ionized in the plasma by resonant charge exchange collisions. The energy is then exchanged to the plasma by coulomb collisions. Barnett[97] points out that one of the serious problems associated with neutral injection is the amount of energetic impurity atoms that enter the plasma. Therefore, in order to tie down the impurity problems charge exchange cross sections are needed for C^+, O^+, N^+, Pt^+, Fe^+, and Ta^+, in H_2, He, and N_2 gas in the energy range from 20 to 300 keV.

For the $D(d,n)^3He$ and $T(d,n)^4He$ reactions energetic ^3He and ^4He ions in the MeV region are produced and therefore charge exchange cross sections are needed for He^{++} and He^+ with atomic hydrogen.

Accelerators can be used in what is called plasma profiling. A heavy ion beam is projected across the plasma, the ion is stripped and becomes doubly ionized. This ionization is dependent on the electron density, proton density, and the neutral density. The stripping cross sections for heavy ions and H^+, H^o and electrons are needed. A rather comprehensive index of other atomic physics needs for the CTR program can be found in Ref. 97.

Many accelerator programs throughout the country are now being devoted to the solution of the problems associated with the mechanisms controlling microstructural changes that take place under prolonged bombardment by fast neutrons and charged particles[108-116]. This general topic is called radiation damage studies. The damage that occurs during irradiation at elevated temperature is mainly in the form of voids which can produce swelling and the acute loss in structural properties of the material. Also for the case of CTR reactors, the (n,p) and (n,α) cross sections in the blanket material are high. These recoiling proton, alphas, and alphas from the CTR, $T(d,n)^4He$ reaction produce gas bubbles and embrittlement effects. Charged particle accelerators are ideally suited to study these phenomena because the damage effects can be enhanced at least three orders of magnitude by bombarding the specimens with heavy ions from an accelerator. Also the damage build up can be controlled and the specimens are not radioactive after bombardment. At Argonne, Taylor et al[108] have designed a facility that will allow simultaneous light ion implantation and heavy ion displacement damage over the same depth range in the sample (approx. 1μM). Figure 11 shows a schematic of this facility.

In addition to damage problems and diagnostics, there is a definite need to assay the possibility of advanced fuels for fusion reactors. The feasibility of $^6Li + D$ has not yet been demonstrated, but there is definitely a need to give consideration to these alternate fuel cycles. McNally from Oak Ridge[117] points out that there are definite needs for accurate cross section measurements for over 80 reaction possibilities. Some of these are shown in Table III. For the reactivity codes used to calculate feasibility, cross sections are needed to an accuracy of 15%.

TABLE III

Charged Particle Nuclear Reactions
For ^{6}Li Fuel Cycle
(Provided by C.F. Barnett[97])

Reaction	Energy Range
$^{6}Li(d,\alpha)\alpha$ + 22.4 MeV	100 keV - 5 MeV
$^{6}Li(d,p)^{7}Li$ + 5.0 MeV	100 keV - 5 MeV
$^{6}Li(d,p)t + \alpha$ + 2.6 MeV	100 keV - 5 MeV
$^{6}Li(d,n)^{7}Be$ + 3.4 MeV	100 keV - 5 MeV
$^{6}Li(d,n)^{3}He + \alpha$ + 1.8 MeV	100 keV - 5 MeV
$^{6}Li(d,d')^{6}Li^{*} \rightarrow d + \alpha$ - 1.5 MeV	3 MeV - 6 MeV
$^{6}Li(\alpha,\alpha')^{6}Li^{*} \rightarrow d + \alpha$ - 1.5 MeV	3 MeV - 12 MeV
$^{6}Li(p,p')\ Li^{*} \rightarrow d + \alpha$ - 1.5 MeV	3 MeV - 15 MeV
$^{6}Li(p,^{3}He)\alpha$ + 4.0 MeV	100 keV - 15 MeV

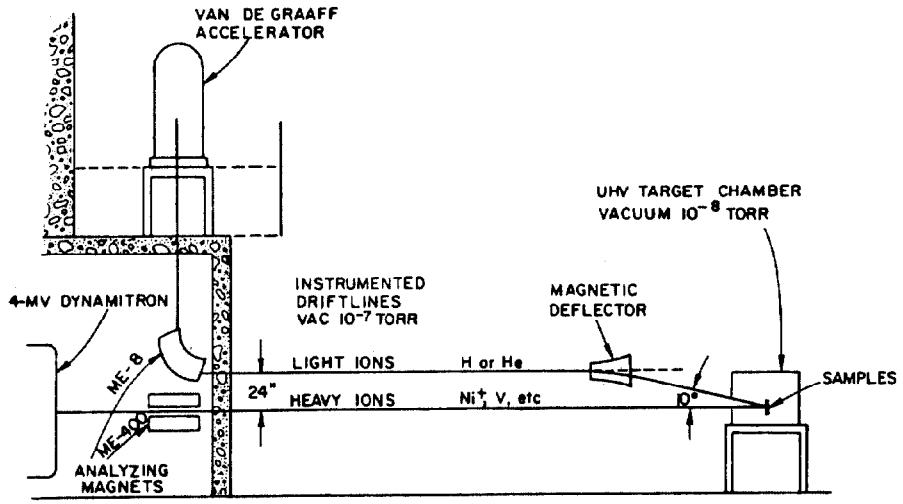


Figure 11. Dual Accelerator System at the Argonne National Laboratory Which is being used to Study Radiation Damage Effects Produced In Reactor Construction Material (Provided by A. Taylor, Ref. 108)

Other articles in regard to these alternate fuel cycles can be found in the literature Ref. 118-122. In summary, the CTR program should continue to supply many challenging problems which require accelerators for a solution.

Other Industrial Applications

There are many other areas of research and industrial applications with small accelerators that are at present receiving a great deal of consideration. For example, the role of the small accelerator in the assay of fissionable materials. These activities are being conducted in conjunction with the ERDA Safeguards Program. Several excellent papers have been written in which these activities are described[123-128].

The small accelerator is also being used extensively for ion implantation in the fabrication of microelectronic devices and circuits. A quick review of the literature[129-135] will show that many new designs of accelerators are coming out to specifically meet the needs associated with this commercial application. Electron-beam-address memory (EBAM) devices are being developed by John Kelly et al.[137-141] at the Stanford Research Institute. These devices have been constructed with data capacities of 1 million bits per tube and random access times of better than 3 μsec.

It was pointed out in the first section of this paper that many accelerators are being used in the medical industry and this number should increase substantially in the next several years. These applications include the use of existing accelerator facilities for analysis and treatment of medical problems[142-149] to the design and fabrication of new machines for specific applications[150-152].

In summary, the small accelerator and accelerator technology is being widely used to solve a multitude of industrial and research problems.

From the vantage point of the authors of this paper, the long range outlook looks good, from both the standpoint of the accelerator design engineer and the individuals who are privileged to use these many fine facilities in Industrial Applications, Research and Teaching.

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