Long-Lifetime High-Yield Neutron Generators using the DD reaction

M. K. Fuller[1], M. A. Piestrup[1], C. K. Gary[1], J. L. Harris[1], G. Jones[1], J. H. Vainionpaa[1], D. L. Williams[1], J. T. Cremer[1], Adam Bell[2], Glenn McRae[2], Daniel Faber[3], B. A. Ludewigt[4], J. W. Kwan[4], J. Reijonen[4], K.-N. Leung[4]

[1]Adelphi Technology, Inc.
[2]Carleton University
[3]Heliocentric Technologies
[4]Lawrence Berkeley National Laboratory

michael.fuller@adelphitech.com

Abstract. The Lawrence Berkeley National Laboratory and Adelphi Technology Inc. have developed a series of high-yield neutron generators using the D-D reaction with an axial geometry. They operate with a single ion beam and can have a small origin size useful for immediate moderation and a high concentration of thermal neutrons. The generator uses RF induction discharge to efficiently ionize the deuterium gas. This discharge method provides high plasma density for high output current, high atomic species from molecular gases, long life operation and versatility for various discharge chamber geometries. These generators are open systems that can be actively pumped for a continuous supply of deuterium gas further increasing the generator's expected lifetime. Since the system is open, many of the components, including the target, can be easily replaced. Pulsed and continuous operation has been demonstrated. In either mode of operation these generators have been used for Prompt Gamma Neutron Activation Analysis (PGNAA) and neutron activation analysis (NAA). Carleton University and Heliocentric Technologies are developing an Elemental Analyzer based on this neutron source.

1. Plasma Neutron Generators

RF plasma neutron generators can produce intense neutron outputs of 10^{10} n/s or greater with the deuterium-deuterium (D-D) reaction and 10^{12} n/s or more with the deuterium-tritium (D-T) reaction. Based on core technology originally developed at Lawrence Berkeley National Laboratory[1] and further developed by Adelphi, these generators provide many times greater neutron output than other portable/transportable systems and have proven to be reliable and easily serviced.

As shown in *FIG. 1*, plasma ion source neutron generators are simple diodes with a plasma source that generates the D+ ions that are then accelerated across a potential of ~100 kV to a titanium target. The target is biased to a negative voltage, while the plasma is at ground. The negative potential will then 'attract' the positively charged particles to the target. The accelerated ions impinge onto a titanium coated copper target where 2.45-MeV D-D neutrons are generated through fusion reactions. The implanted hydrogen ions form titanium hydrates in the titanium matrix, thus 'trapping' the deuterium atoms. When the generator is operating, the titanium layer is initially being loaded with deuterium and the neutron output is increasing with time; then after some minutes of operation the target is loaded by the incoming ions and the neutron output saturates to a stable level. While the ions are implanted to the target, heat is also deposited, which is removed from the target by cooling water.

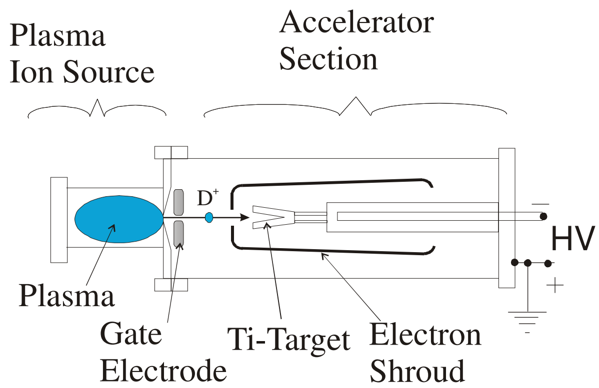

FIG.1. Simple diagram of the principle components of the neutron generator using the D-D reaction.

1.1. Current Generators

Adelphi Technology produces high intensity, neutron generators based on this technology. Adelphi currently makes devices with outputs of 10^8 (DD-108), 10^9 (DD-109) and 10^{10} (DD-110) fast neutrons per second. The model DD-109 is shown in Fig. 2. In addition, Adelphi has built a neutron generator specialized for the production of thermal neutrons for both prompt and delayed gamma neutron activation studies. These generators evolved from work

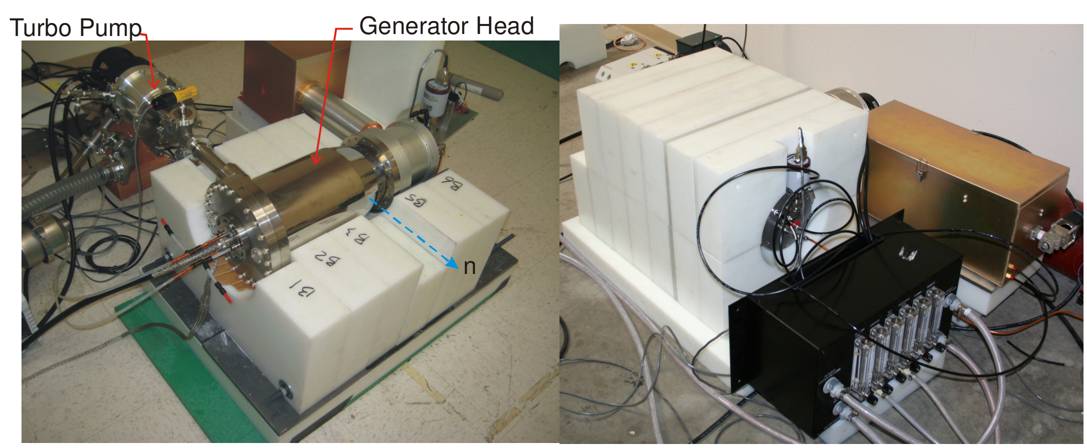

FIG. 2. The model DD-109 neutron generator head shielded inside a polyethylene housing.

sponsored by the Department of Energy to develop a source for non-destructive testing at nuclear power plants [2]. The current products are all continuous-duty (non-pulsed) neutron generators using the deuterium-deuterium (D-D) fusion reaction.

The neutron flux depends on the ion current and acceleration voltage. The voltage is easily controlled, and the current can be changed by varying the plasma density (both by varying the excitation and gas density). This allows continuous and precise control of the neutron output, and if the output is measured, a feedback loop can maintain a constant output flux over extended times. Because the voltage, gas pressure and RF excitation can be computer controlled, the neutron generators have been equipped with software to allow their control remotely and even over an internet connection. Generators using the D-D reaction have the further advantage that the system can be opened and serviced, greatly increasing its lifetime.

1.2. Generator Yield

The yields of the three models have been measured for both continuous and pulsed operation. With one exception the measured yields are close to the expected calculated values. The DD-108 and DD-109 have each been measured at the design values of 10^8 n/s and 10^9 n/s. At present the prototype DD-110 is operating at 1.8 x 10^9 n/s, below its desired output of 10^{10} n/s. The present extraction iris area is smaller than that required for high yield production. The aperture area determines the ion beam current. At present the area limits the beam current to 15-18 mA. Larger irises are expected to achieve the desire yields. As shown in Fig. 3, the expected yield per mA is close to the desired calculated value. Pulsed operation is

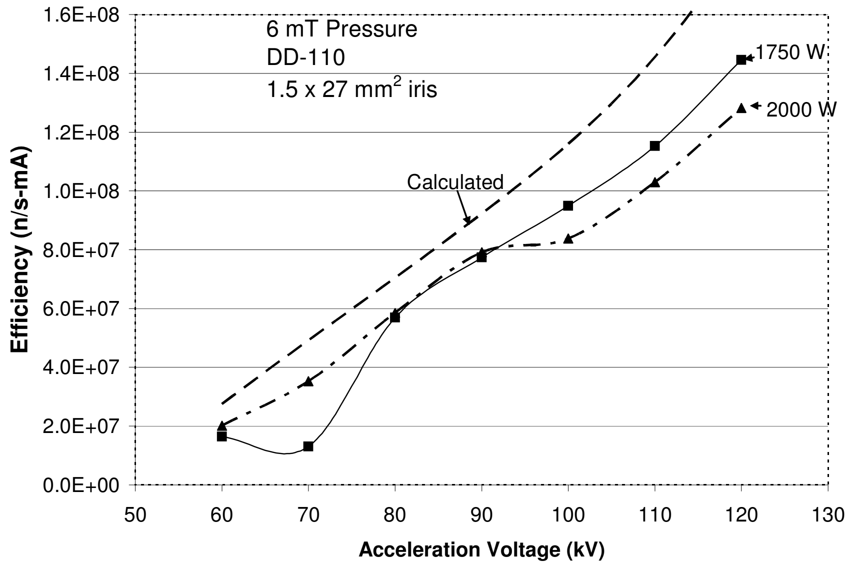

FIG. 3. The efficiency of neutron production per mA of beam current as a function of accelerator voltage for two RF plasma powers.

achieved by pulsing the 13.56 MHz RF supply to the plasma ion source. Pulse lengths greater than 100 µs can be achieved.

2. General Applications

While the intensity of older-technology Penning diodes is sufficient for oil well logging, the greater output offered by Adelphi's plasma sources offers a larger range of laboratory research opportunities, including neutron activation studies, explosives detection, detection of special nuclear materials, neutron radiography, and chemical analysis.

Detection of Conventional Explosives: Neutron interrogation is a valid method for determining the presence of concealed conventional explosives. There are several different techniques used, with effectiveness depending on the particular application. Whether thermal activation (measuring prompt or delayed gammas), fast neutron resonance/excited emission, or neutron radiography are used, increased flux will increase the range of detection – a critical parameter. Some of these projects have benefited from Adelphi high-output source technology. The research efforts should yield a viable solution for shipping containers in the next few years.

Special Nuclear Material (SNM) Detection: Today, the detection and verification of SNM is an important challenge for the world community. As rogue states continue to develop and acquire nuclear weaponry, deterrence against use will employ new strategies and tools. While advances in passive detection systems will enhance the probability of detecting non-shielded or weakly shielded radioactive materials, the detection of shielded SNM and in particular shielded Highly Enriched Uranium (HEU) poses a significant challenge that is best addressed using active detection systems. These systems can be utilized to inspect cargo in shipping containers at seaports, border crossings, and air transport containers, or to be deployed as mobile inspection systems. For the pulse die-away technique for locating SNM, researchers have shown that it is important to radiate with neutrons below 10 MeV[7,8] from a pulsed source. For example, there is an important interference line from ^{16}O by the $^{16}O(n,p)$ ^{16}N reaction that produces a strong 6-MeV γ-ray line [8]. This interference is important when irradiating with 14-MeV neutrons from the D-T reaction since the reaction threshold neutron energy is 10 MeV. By operating with the D-D reaction, we can produce a neutron beam with energy of approximately 2.5 MeV. Adelphi has already attained the necessary pulsing regime.

Chemical Industry: PGAA and NAA are established tools for elemental analysis and can potentially provide a complete picture of all contaminants in inorganic chemicals. Importantly, many applications require a higher neutron flux than currently possible with economically viable (much less than $1M) sources.

Semiconductor Industry: Impurities in semiconductor materials may have a serious impact on the efficiency of electron transport throughout the chip matrix. In addition, sometimes there may be a need to dope the materials with infinitesimal amounts of a particular element. Contaminants are easily detected in inorganic matrices using PGAA/short-lived NAA. Having this level of analytical detail may offer insights into improving the semiconductor manufacturing or monitoring the doping processes.

3. Specific Application in a Neutron Activation Analysis (NAA) Mining Instrument

Heliocentric Technologies, working with Carleton University, is developing an Elemental Analyzer. This device uses Prompt Gamma Neutron Activation Analysis (PGNAA) to determine the elemental composition of samples with no prior sample preparation. The device is designed to be compact while still accommodating samples that can weigh up to a few kilograms.

A key component of the prototype Elemental Analyzer is an accelerator-based D-D Neutron Generator, which provides a high neutron flux for irradiating samples. Most commercially available PGNAA systems use either research reactors or high yield, neutron-emitting radioisotopes. The accelerator-based neutron generator provides the ability to throttle the intensity of the neutron field, or even terminate or pulse the neutron production on command. This is a huge advantage over other neutron sources in term of safety (even the fuel source is not radioactive), serviceability, flexibility and mobility.

The other system components are the radiation shielding and neutron moderator, a High-Purity Germanium (HPGe) semiconductor photon detector with electronics, and the data processing algorithms. *FIG. 5* shows the pre-production concept of Elemental Analyzer.

3.1. Instrument and Test Sample Description

Heliocentric's Elemental Analyzer is currently in the advanced prototype phase of design. A series of tests was performed in November 2008 in order to validate the prototype's performance targets. A set of calibration samples, as well as representative samples from various customers, were used to assess the prototype's performance.

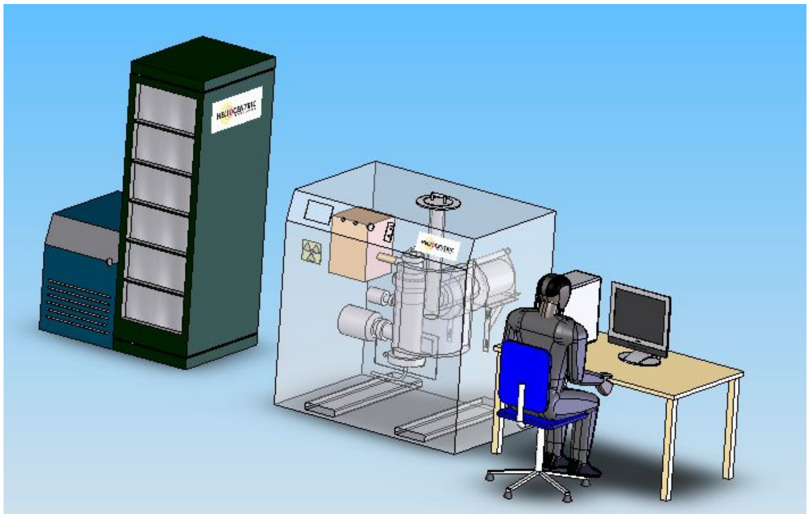

FIG.5. Pre-Production concept of the Elemental Analyzer. The electronics rack and cooler (left), the Adelphi neutron generator, moderator/shielding and detector (center, translucent), command station and operator (right).

The neutron generator supplied by Adelphi Technology was the Model DD-109. The device uses a deuterium-deuterium reaction to produce large quantities of 2.45 MeV mono-energetic neutrons. The neutron yield of the generator was on the order of 10^9 n/s isotropic. The average flux in the test sample is estimated to be 10^5 $n \cdot cm^{-2} \cdot s^{-1}$. The test chamber (approximately 1.5 L volume), where the test samples are placed, was surrounded by a moderating material to optimize the thermal neutron flux within the sample. A shielding material surrounded the Analyzer components in order to protect the operators from the radiation and to limit the incursion of background radiation.

When the nucleus of an element becomes activated by thermal neutrons it will emit photons of characteristic energies. A coaxial n-type HPGe detector (~ 100 cm^3 crystal volume) was used to detect that characteristic radiation. A digital Multi-Channel Analyzer (MCA) was used to record and bin the detector signals.

The data from the MCA is simply a spectrum containing the number of counts registered over a range of photon energies. Calibration of the Elemental Analyzer allows the software to convert numbers of counts to grams of material, with further processing to accommodate samples of a variety of compositions and physical and material properties.

Calibration samples consisted of pure elements and ranged in size from 1 to 64 g. These samples were used to establish a relationship between the number of counts registered by the detector and the mass of the sample present in the Analyzer test chamber. The customer samples that were tested include mining rock samples, powder, granules and irregularly-shaped "lump samples". *FIG. 6* contains examples of the mineral samples.

(a) (b) (c)

FIG. 6. Examples of mining samples tested in Heliocentric's prototype Elemental Analyzer. Split rock samples (a), Powder (b) and "lump samples" (c).

3.2. Results of Testing

The primary purpose of this series of tests was to verify the scalability of the design concept by validating the prototype's target detection limits and measurement accuracy. Previous tests had been conducted with a neutron source 1000 times weaker than the accelerator-based neutron generator. These tests allowed an assessment of the impact of those differences on the system performance.

Calibration samples were loaded into the test chamber and a photon spectrum was recorded while the samples were simultaneously bombarded by neutrons from the neutron generator for 1000 seconds (live time, not real time). The PGNA spectra from the calibration samples were post-processed to produce calibration curves for each individual element (*see FIG. 7*). The relationship between count rate and sample mass is necessary to interpret the customer sample spectra.

The customer samples experienced the same test procedure as the calibration samples. The post-processing algorithms calculated the quantity of material present in each of the samples using the calibration data and accounting for each individual sample's unique physical and material properties. The compositions of the customer samples measured with the prototype Elemental Analyzer were compared with the known sample compositions provided by the sample suppliers to assess the Elemental Analyzer's accuracy (*see FIG. 8*). Improvements to accuracy can be obtained through longer test times for the calibration samples; time constraints on testing prevented such considerations in this case.

The detection limit (DL) has been defined as the level at which a measurement has a +/-100% precision for a pure calibration sample. For example, if the DL is 5 grams and a 5 gram calibration sample is tested, over many tests the average result will be 5g with a standard deviation of 5g. The accuracy of the instrument is such that all measurements above the DL will be within 15% of the true value 95% of the time.

Detection limits for the Elemental Analyzer were calculated for eight elements of interest (*see TABLE 1*). The current Elemental Analyzer is a prototype designed to validate intermediate scalability of the design. The final, production-ready instrument will require further scaling in terms of shielding, data processing and detector technology. The final estimated detection limits are also shown in TABLE 1.

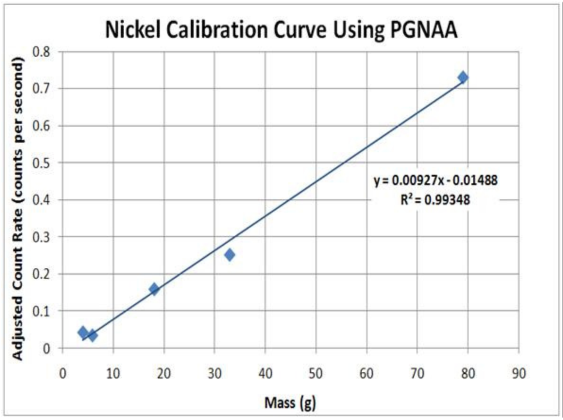

FIG. 7. *Calibration curve for pure Nickel. The count rate has been adjusted to account for sample physical properties.*

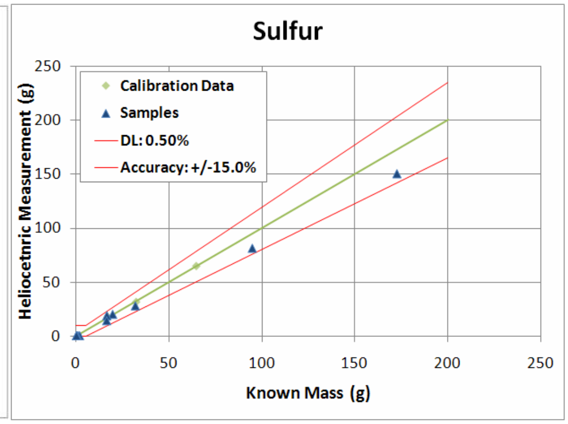

FIG. 8. A graph comparing measured mass versus the known sample mass for sulfur in customer samples. All measurements are within +/- 15% of the actual values.

TABLE 1 – DETECTION LIMITS FOR HELIOCENTRIC'S ELEMENTAL ANALYZER

Element		**Detection Limit: prototype**	**Detection Limit: target for final instrument**
Aluminum	Al	0.2% (Delayed GNA)	0.02 % (Delayed GNA)
Arsenic	As	N/A	0.1 %
Cobalt	Co	0.5%	0.05 %
Copper	Cu	0.6%	0.1 %
Chlorine	Cl	N/A	0.05 %
Chromium	Cr	0.9%	0.1%
Iron	Fe	1.5%	0.1%
Nickel	Ni	0.5%	0.05 %
Silver	Ag	N/A	0.01 %
Sulfur	S	0.5%	0.1 %
Zinc	Zn	3%	0.2 %
Uranium	U	N/A	0.01% (DNC/PFN)
Integration time	t	1,000 s	300 s
Absolute accuracy	+/-	15%	5%

3.3. Deduction from Prototype Testing

The compact Elemental Analyzer being developed by Heliocentric Technologies employs PGNAA to quickly and accurately determine the elemental composition of samples without any sample preparation. It has been demonstrated that the Elemental Analyzer, incorporating a D-D neutron generator with 10^9 isotropic output, can achieve the target detection limits and accuracy for the current prototype between 5 and 15 grams (0.5% to 1.5% in 1kg samples).

During a series of tests in November 2008, the prototype Analyzer was able to provide the elemental compositions of an array of samples including Minerals, Powder, Granules and various "lump samples". These measured elemental compositions were within the acceptable range of error of the known sample compositions. Future work includes refinements to the design and algorithms to allow the instrument to achieve the target performance for the final production instrument: detection limits between 0.05 and 0.1 grams for the elements of interest.

4. Conclusion

RF plasma neutron generators can produce intense neutron outputs with the deuterium-deuterium (D-D) reaction. Because the energy of the neutrons produced, moderation to thermal energies can be achieved in reasonable geometries. This capability offers a large range of laboratory research opportunities and industrial applications. A particularly promising application is for the elemental composition analysis of rock core samples.

Appendix 1: References

[1] J. Reijonen, N. Andresen, F. Gicquel, R. Gough, M. King, T. Kalvas, K.-N. Leung, T.-P. Lou, H.Vainionpaa, A. Antolak, D. Morse, B. Doyle, G. Miller and M. A. Piestrup, "Development of Advanced Neutron/Gamma Generators for Imaging and Active Interrogation Applications," Proceedings of SPIE Vol. 6540-55 (SPIE, Orlando, FL, 2007).

[2] J. T. Cremer, Principal Investigator DOE Phase II STTR (DE-FG02-04ER86177), "High Brightness Neutron Source for Radiography," (7/13/05-07/10/2007).

[3] M. A. Piestrup, Principal Investigator Phase I STTR (DE) "Gamma Ray Calibration Source," DE-FG02-07ER86294.

[4] J. W. Kwan, R. Gough, B. A. Ludewigt, M. Regis, R. P. Wells, J. H. Vainionpaa, "A 2.45 GHz High Current Ion Source for Neutron Production," High Energy Physics and Nuclear Physics, (2007).

[5] J. H. Vainionpaa, R. Gough, M. Hoff, J. W. Kwan, B. A. Ludewigt, M. J. Regis,J. G. Wallig, R. Wells, "Microwave ion source and beam injection for an accelerator-driven neutron source," Proceedings of PAC07, Albuquerque, New Mexico, USA TUPAS045 (2007).

[6] Qing Ji, Y. Wu, M. Regis, J. W. Kwan, "Compact Portable Microwave Driven Neutron Generator, LBNL (2008).

[7] G J Roach and J R Tickner "Performance Tests on Adelphi Neutron Generator Carried out during Overseas Visit 18-22 June 2007" CSIRO Report DMR-3252 (2007).

[8] D. Slaughter, M. Accatino, A. Bernstein, J. Candy, A. Dougan, J. Hall, A. Loshak, D. Manatt, A.Meyer, B. Pohl, S. Prussin, R. Walling, D. Weirup, "Detection of Special Nuclear Material in Cargo Containers Using Neutron Interrogation", Lawrence Livermore National Laboratory, UCRL-ID-155315, August 2003.