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AN EVALUATION OF SEALED-TUBE NEUTRON GENERATORS FOR THE ASSAY OF FRESH LWR FUEL ASSEMBLIES

by

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ABSTRACT

The use of sealed-tube neutron generators for the active assay of fresh light-water reactor fuel assemblies has been investigated. The results of experimental tests of the Kaman 801 generator are presented. Neutron yields, source moderation, and transportability are discussed. A comparison is made with the AmLi neutron source for use in the Coincidence Collar.

I. INTRODUCTION

An important safeguarding task identified by the International Atomic Energy Agency (IAEA) is the nondestructive verification of the fissile content in unirradiated light-water reactor (LWR) fuel. During the past several years, two portable active neutron interrogation systems for LWR fuel assemblies have been developed by the Safeguards Assay Group (Q-1) at the Los Alamos National Laboratory. These devices use AmLi sources as the interrogating neutron source. However, because it is difficult to transport radioactive sources, neutron generator tubes are now being considered as alternative neutron sources to alleviate some of the transportation and licensing problems.

Neutron generators¹ have been used for several years by the uranium well-logging industry to assay the quantity of uranium underground in exploration

boreholes. Most of the well-logging systems have used portable neutron generators to supply the interrogation neutrons needed for measuring the ^{235}U content in the ore. The logging need has led to the development of compact and rugged neutron generators that can be used in relatively small boreholes under several thousand feet of water. The environment for fresh fuel assembly measurement is much more favorable.

Pulsed neutron generators together with the prompt neutron signal have been used by Sandia National Laboratories, Albuquerque,² for the measurement of fissile material in a portal monitor and by the Detection and Verification Group (Q-2)³ at Los Alamos for the assay of small quantities of ^{235}U in large waste containers. Both of these systems require large quantities of graphite and CH_2 moderators to extend the lifetimes of the thermal neutrons in the system, which increases the sensitivity for small quantities of fissile material.

At the request of the IAEA (Task A.31), Los Alamos has conducted a study⁴ of the use of a neutron generator as a replacement for radioactive sources. This report details the results of that study and gives an experimental evaluation of several neutron generator systems.

In addition to the present safeguards application, neutron generators have been evaluated at Los Alamos for the measurement of nuclear materials in large waste containers. A summary of the experience of using the Sandia MA-165 sealed tube for this work is given in the Appendix.

II. EXPERIMENTAL METHOD FOR LWR ASSEMBLY VERIFICATION

The verification of LWR fuel assemblies requires the measurement of ^{235}U content in full fuel assemblies. Two active neutron instruments have been developed for this purpose: the Neutron Collar and the Coincidence Collar.

A. Neutron Collar

The first unit developed several years ago is called the Neutron Collar.⁵ It consists of a polyethylene main frame, an $^{241}\text{AmLi}$ neutron source, two ^4He detectors, and an electronic counting system. The Neutron Collar without the processing electronics is shown in Fig. 1 and the basic characteristics of the system are described in Ref. 5.

Fig. 1.
Diagram of the Neutron Collar.

The AmLi source neutrons have a distribution of energies with a peak at ~ 0.3 MeV; very few neutrons have energies greater than 1.0 MeV. The polyethylene between the source and the fuel assembly moderates the source neutrons to thermal and epithermal energies, thus ensuring that the induced fissions in the fuel assembly are primarily from ^{235}U and not ^{238}U . This technique is commonly called subthreshold interrogation because most of the neutron energies are below the ^{238}U fission threshold.

The mode of operation of the Neutron Collar is the prompt mode; that is, the ^4He neutron detectors detect the prompt fission neutrons from the fuel assembly while the neutrons from the neutron source are inducing fissions in the assembly. The energy thresholds for the ^4He detectors are normally set to be insensitive to the lower energy interrogating source. A Silena multi-channel analyzer (MCA) is normally used as the support electronics.

A recent test of the Neutron Collar in an in-plant situation demonstrated that the instrument is capable of a relative measurement of ~1% precision (1σ) at a count time of 1000 s.

B. Coincidence Collar

Because the Neutron Collar gave poor penetration to the central regions of the fuel assembly and sampled only one side of the assembly, a Coincidence Collar was developed.⁶ This instrument uses neutron interrogation with an AmLi neutron source and coincidence counting of the induced fission reaction neutrons from ^{235}U . The coincidence counting separates the fission neutrons, which originate from ^{235}U , from the random neutrons used in the interrogation. This approach has the advantage over that used for the Neutron Collar in that the source strength is a factor of 10 smaller, and the sensitivity for interior fuel pins is better.

The Coincidence Collar consists of three banks of ^3He tubes and an AmLi source imbedded in a high-density polyethylene (CH_2) body, as shown in Fig. 2. The 18 ^3He neutron detector tubes (4 atm pressure) are 2.54 cm in diameter and 33 cm long (active length).

The CH_2 body performs three basic functions in the system: (1) general mechanical support, (2) interrogation source neutron moderation, and (3) slowing down of induced fission neutrons prior to their detection in the ^3He tubes.

C. Neutron Generator Interface

Replacement of the AmLi source by the neutron generator has different constraints for the two different types of collars described above. The Neutron Collar uses subthreshold

Fig. 2.

Diagram of the Coincidence Collar showing the AmLi neutron source and the ^3He detector banks. The top detector bank pivots open to accommodate PWR, BWR, or HWR fuel assemblies.

irradiation and fast neutron integral counting, whereas the Coincidence Collar counts coincidence neutrons. The corresponding constraints are outlined below:

1. Neutron Collar

- a. Requires a (D,D)* neutron generator to give subthreshold irradiation for low detector background rate.
- b. Uses either pulsed or direct current mode of generator operation.
- c. Needs a separate neutron source flux monitor to correct for neutron source yield fluctuations.

2. Coincidence Collar

- a. Uses either (D,D) or (D,T) neutron generators.
- b. Cannot use pulsed mode of operation because of coincidence interference.
- c. Does not need a separate neutron flux monitor, because of totals rate information.

It is possible to operate the Coincidence Collar in the integral (non-coincidence) mode by counting the induced delayed neutrons from a pulsed source rather than the prompt coincidence neutrons. However, this requires (1) additional electronics to gate the electronics off during the interrogation pulse and (2) a separate source flux monitor.

In all cases it is necessary to have one set of electronics to operate the neutron generator system and an additional set to operate the detector system.

III. NEUTRON GENERATOR DESCRIPTION

A. Neutron Yields

Small sealed-tube neutron generators are commercially available at two neutron energies: ~14.3 MeV from the (D,T) reaction and ~2.5 MeV from the (D,D) reaction. The neutron energy most compatible with a portable system depends on the neutron yield from each reaction, source moderation requirements, the type of detector, personnel radiation safety, and transportability.

The theoretical thick-target yields for the (D,D) and (D,T) nuclear reactions have been calculated for the deuteron energy range 0-300 keV⁷ and are

*Notation of (D,D) refers to the $D(D,^3\text{He})n$ reaction, and (D,T) refers to the $D(T,^4\text{He})n$ reaction.

shown in Fig. 3. At 150 keV, the (D,D) yield is more than 2 orders of magnitude less than the (D,T) yield. The (D,T) reaction is isotropic, that is, the neutrons are emitted uniformly in all angles; however, the (D,D) reaction is anisotropic and has an angular dependence that is peaked in the forward direction. For comparable accelerating potentials and beam currents (corresponding to comparable target heating), the differences in yield in the forward direction between the two reactions is approximately 2 orders of magnitude.

B. Neutron Moderating Material

To enhance the ratio of fissile response to fertile response, both types of neutron generators require neutron spectrum tailoring; however, the (D,T) generator requires considerably more. For LWR fuel assemblies,

the ratio of fertile material (^{238}U) to fissile material (^{235}U) varies from ~30/1 to 50/1. Because the induced fission cross sections are comparable for ^{235}U and ^{238}U above 1 MeV, the fissile response due to unmoderated source neutrons (either 2.5 or 14.3 MeV) can be masked by the fertile response. The fissile-to-fertile response can be enhanced by shifting the energy of the interrogating neutrons to below the threshold (~1 MeV) for the fission of ^{238}U . Spectrum tailoring must also be employed to reduce the energy of the neutrons to below some reasonable detector bias level so that prompt fission neutrons can be detected above the background resulting from the interrogating source neutrons. The Coincidence Collar moderates the AmLi source neutrons to thermal and epithermal values through the use of polyethylene. It is expected that a moderately thin polyethylene configuration can be designed for the (D,D)

Fig. 3.
Theoretical thick-target yields for the (D,D) and (D,T) nuclear reactions.

generator to provide a satisfactory fission ratio of $>100/1$ for the LWR fuel assemblies. A thicker moderator will be required for the (D,T) generator.

C. Neutron Generator Tubes

Generally, the commercially available neutron generator systems have been custom designed for applications such as logging. It is likely that a custom-designed or slightly modified system will be required for application to both types of neutron collars. Table I lists some of the sealed tubes that are available.

Considerations of cost and availability limit the choices to the neutron generators marketed by Kaman Sciences, Inc. These tubes have an advertised lifetime in excess of 200-400 h. For portability purposes, target cooling is not acceptable, so that the limitation on the operation of both tubes is target heating, with the neutron yield limited to 10^8 - 10^9 n/s (D,T) or 10^6 - 10^7 n/s (D,D). At these yields, both types of tubes are comparable, and a choice between the two is made on size considerations and ease of electrical connections. The A3041 tube is smaller and simpler to connect electrically. The Zetatron tube and transformer assembly has been used successfully in borehole logging and waste assay measurements as described in the Appendix.

General Electric has produced a number of tubes that have been used at Sandia¹ and Los Alamos,³ these tubes have been recently made available for commercial procurement from Kaman Sciences, Inc.

For the present work, we have concentrated on (D,D)-type tubes because of the low yield ($<10^7$ n/s) and low-energy neutron requirements and a desire to avoid the radioactive material (that is, tritium) in (D,T) tubes. A typical (D,T) tube contains on the order of 10 Ci of tritium.

IV. EXPERIMENTAL TESTS

Figure 4 shows the general electronic configuration of our experimental tests on the Kaman Sciences, Inc. Model 801 sealed-tube neutron generator. The experimental configuration is explained in the following two sections. An evaluation of (D,T)-tube models by Los Alamos is described in the Appendix. A comprehensive description of the (D,T) Sandia Zetatron device can be obtained in Ref. 8.

TABLE I

SEALED-TUBE NEUTRON GENERATORS

<u>Manufacturer</u>	<u>Tube Type</u>	<u>Mode</u>	<u>Neutron Yield (D,T) (n/s)</u>	<u>Acc. Voltage (kV)</u>	<u>Approximate Size (cm)</u>	<u>Comments</u>
Kaman Sciences, Inc.	A3041	Pulse or dc	$>10^8$	+100	7 by 17	Primarily pulsed, cost \$4-6K
	A3043	Pulse or dc	10^{10} a	-150	9 by 22	Primarily used dc, cost \$6-8K.
	Zetatron	Pulse	1×10^8	---		Used for logging 10K
Marconi Avionics	K type	Pulse or dc	10^8	-120	5 by 20	6-9 months delivery time
Haefely ^b 12 months delivery		Pulse or dc	5×10^{12} a		+200	36 by 58.5 time, cost \$500K
Schlumberger	Neutron	Tube not available commercially				
General Electric ^c	Neutron					

^aTubes in excess of 10^9 n/s normally require target cooling.

^bUnder license agreement with the Center for Nuclear Research at Karlsruhe, West Germany.

^cTube developed for Sandia National Laboratories, now under license agreement with Kaman Sciences, Inc., for commercial production.

Fig. 4.
Logic diagram for the (D,D) neutron generator test setup.

A. Detection System

The detection system consisted of the sealed-tube neutron generator, the polyethylene moderator, ^3He tubes, and a scintillator. The detection system configuration is shown in Fig. 5.

The sealed tube was based on a (D,D) reaction, using a scandium deuteride target, producing up to 2.5-MeV neutrons. The tube was operated in the pulsed mode. The manufacturer's rating of the tube is approximately 10^6 n/s with a pulse width of 2.5 μs , and a pulse rate of 1-10 pps.

Fig. 5.
Schematic diagram of the experimental setup for evaluating the neutron generator with the Neutron Collar.

Polyethylene was used as a moderator for the neutron source and the ^3He tubes. The polyethylene was 2.5 cm thick between the ^3He tubes and the fuel assembly and between the neutron generator and the fuel assembly. The ^3He tubes were divided into two groups of four as shown in Fig. 5. All the tubes are 2.54 cm in diameter, and the polyethylene covered the bottom 30 cm of the tubes. The voltage on the ^3He tubes was 1450 V.

The scintillator was an NE213 liquid. The shape was that of a 7.6-cm by 7.6-cm right circular cylinder, attached to an RCA 8575 photomultiplier tube with an Ortec 265 phototube base. The photomultiplier was operated at 1500 V.

B. Electronic Hardware and Software

The electronics consisted of the neutron generator high-voltage power supply and a microprocessor-based data acquisition system.

The high-voltage pulsed power supply was a Kaman Model 801 control console weighing approximately 77 kg and measuring 55 by 50 by 55 cm. An automatic gas reservoir control maintained the desired deuterium gas pressure in the sealed tube. The delayed neutron detection system's electronics consisted of an Ortec 142 PC preamplifier and a Canberra 2015-A amplifier/single-channel analyzer, which provided a logic pulse for the scaler.

The microprocessor data acquisition system utilized a Motorola MC6800 microcomputer with a 1-MHz internal clock. The microprocessor contained 8K of RAM and 10K of ROM. The microprocessor also contained an analog-to-digital converter (ADC) card and a scaler card. The ADC card converted and recorded the scintillator signal and the scaler card counted the ^3He tube signals.

The ADC card was a custom-made interface card that integrated the current signal from the photomultiplier tube and digitized the analog level with a 256-channel bipolar ADC. The ADC card was controlled by the microprocessor. A schematic of the ADC card is shown in Fig. 6.

The scaler card was also a custom-made interface card that was designed to operate in a variety of configurations. For this experiment, the scalars were operated in the time-interval mode, that is, the scalars were active for a preset time, set by the software. The maximum preset time was 10 s and the maximum count rate was 100 kHz. The scaler card had a capacity of over 65K counts. A schematic of the scaler card is shown in Fig. 7.

The experiment was operated under software control. Virtually all delays, pulses, and their durations were controlled by the software. Figure 8 shows a

Fig. 6.

Circuit diagram of the interface card for integrating the scintillator signal.

timing diagram of the experiment. Each pulse cycle started with a reset pulse. At the same time that the integrator was reset, a signal was sent to an external delay generator. The external delay generator provided the experimenter with the flexibility required to synchronize the gate and the firing of the neutron generator. This was necessary because the gate had to be closed following the neutron generator pulse so that the scintillator pulse could be integrated by the ADC card. Due to saturation of the preamplifier in the first 30 μ s after firing, a 30-ms delay was introduced before the scalers were started. The scalers were run for 1 s. The duration of each cycle was variable so that different repetition rates were possible.

By using a microprocessor, the experiment was made very flexible. Quick changes in the program were relatively easy because the software mode allowed

Fig. 7.

Schematic diagram of the scaler card used in the sealed-tube test setup.

Fig. 8.

Trimming gates used in the control of neutron generator pulsing.

changes in delays, pulse width, and even the mode in which the scalers were run. In fact, program changes were made between runs as new data became available that indicated changes were needed in the experiment.

V. TEST RESULTS

A. Characteristic Operation

Daily preparation and operation of the neutron generator varied considerably from the initial operation to the final run. After a 5-min warm-up period, it was possible to produce neutrons in the initial runs in a relatively

short period of time, ≤ 5 min. However, it was not possible to use the gas reservoir current settings from a previous day to obtain satisfactory operation; manual adjustments were always necessary. In each succeeding run, the reservoir current had to be increased from the previous settings. The gas reservoir failed due to lack of gas during the final run and neutron production ceased. During the later stages of testing, 1000-3000 pulses at reduced target voltage were necessary before the sealed tube operated properly and produced neutrons. The behavior of the tube indicated that the reservoir was gradually being depleted of the deuterium gas at a rate far in excess of that expected. This could have been due to gradual absorption of the deuterium gas by an improperly fabricated target. Near the end of tube life, ~ 1 h (3600 pulses) was required to initiate neutron production.

B. Short-Term Pulse-to-Pulse Stability

Short-term stability was determined by integrating the prompt fast-neutron signal from the liquid scintillator for each pulse of the neutron generator and comparing this over a 1000-pulse run. The pulsing rate was 1 pps. A plot of the results for a typical run is shown in Fig. 9. We plotted the integrated neutron signal for 100 pulses at the beginning, middle, and end of a 1000-pulse run. Except for the last three runs, the average pulse-to-pulse stability was less than $\pm 5\%$. Two factors contributed to this 5% instability and are clearly evident in Fig. 9. The first 70-80 pulses were unstable and had an occasional misfire in which the neutron yield was abnormally low. The instability was reduced to less than $\pm 2\%$ when the first 100 pulses were ignored. Three runs were made with the voltage to the sealed-tube transformer reduced from 4.5 kV to 3.8 kV. These runs also exhibited some initial pulse instability, but even with all the points (1000) included, the pulse instability was $\pm 1\%$. No occasional misfiring was observed.

C. Long-Term Stability: Delayed Neutron Measurement

The delayed neutron yield from a boiling-water reactor (BWR) fuel assembly was used as a measure of the long-term stability. A series of measurements was performed until we could no longer operate the sealed tube; the pulse rate was 1 pps. A plot of these runs is shown in Fig. 10. We plotted the integrated yield over a 1000-s run and, in addition, subdivided the results by integrating from 100-500 pulses and 100-1000 pulses. By eliminating the first 100 pulses,

Fig. 9.
Integrated neutron signal results for the beginning (bottom plot), middle (center plot), and end (top plot) of a 1000-pulse run.

Fig. 10.
Integrated delayed neutron yield from a BWR mockup
assembly over a 1000-s run.

we hoped to minimize the effect on the assay of the instability of the first 70-80 pulses. Three additional runs with the high voltage reduced from 4.5 kV to 3.8 kV are included in the plot. The standard deviation for the 4.5-kV runs was 8.5%; with the last three unstable runs excluded, the error was 7%. For the limited set of three 3.8-kV runs, the variation was 1%.

By normalizing the delayed neutron yield with the integrated scintillator signals, the measurement was improved. However, for the measurement reported here the scintillator response appeared to be saturated, so that the normalization procedure did not adequately remove the dependence of the measurement on the neutron generator yield.

VI. CONCLUSIONS

A. Electrical Safety

The pulse neutron generator, Kaman Model 801, is a transportable device suitable for in-plant installation and designed to be electrically safe under normal operating conditions. However, since potentially lethal voltages are present in the high-voltage power supply electronics cabinet, personnel should be trained in the safe operation of the device and only technically qualified personnel should perform repair or maintenance.

B. Reliability

The high-voltage power supply electronics had electrical failures in three reservoir current transformers, the pulse counting network, and the automatic pulsing network. These failures raise serious questions concerning the reliability and quality control of the electronics. The reservoir current transformer was replaced with a higher power rating transformer so that sufficient power was available to operate the sealed tube.

When the sealed tube was in operation, assays of $\pm 5\%$ were possible. As stated before, pulse instabilities occurred during the first 100 pulses. During this period of operation it was not possible to use the reservoir settings from one day to the next and maintain satisfactory operation. Manual adjustments were always necessary in order to initiate neutron production. Eventually, the sealed tube ceased adequate operation, long before the manufacturer's specifications claimed and the warranties expired. This suggests that the

system suffered from a failure in fabrication, which could explain the lack of reproducible operation from one day to the next.

A second tube was built by Kaman to replace the failed one. It operated successfully for approximately 10 000 pulses. The operation of this tube was similar to the first. The gas reservoir required increasing amounts of electrical power each day that the tube was operated. For both tubes, the amount of power required near the end of the tube life was approximately 55-60 W. At the beginning of the tube life, we used 4 W of power to liberate sufficient quantities of deuterium gas from the titanium reservoir for proper gas pressure.

C. Operation Mode

The tests were performed with neutron generators operating in the pulsed mode. This was done to reduce high-voltage requirements in the power supply so that the unit would be transportable. Direct-current tubes require more power, and the power supplies have an oil-bath insulation with a weight of roughly 45 kg for the ~100-kV power supplies. This extra weight reduces the portability of the unit.

The Coincidence Collar currently used by the IAEA uses the shift register electronics for counting coincidence neutrons from ^{235}U . Because a pulsed neutron source emits copious quantities of time-correlated neutrons, it is incompatible with the coincidence counting technique.

D. Assay Precision

The neutron generator gave a neutron-yield reproducibility of roughly $\pm 5\%$ (1σ). This variation could possibly be reduced to $\pm 1-2\%$ if normalized to a neutron-yield monitor. Table II shows the effect of this type of variation on a pressurized-water reactor assembly measurement and compares the AmLi source now used by the IAEA with the neutron generator tubes.

E. Summary

In summary, the lack of reliability of the Kaman Model 801 sealed-tube neutron generator and its poor precision, complexity, and electrical safety problems make it a second choice to the AmLi source now in use. As the new generation of neutron sealed tubes becomes commercially available, improvements are expected. The technology should be periodically reviewed for ways of

taking advantage of the shorter measurement times and simpler licensing procedures that a neutron generator might offer.

The favorable test results described in the Appendix indicate that certain types of sealed-tube neutron generators can give satisfactory results when operated by skilled technicians in a laboratory setting.

TABLE II

EFFECT OF NEUTRON STABILITY ON FUEL ASSEMBLY ASSAY

	Precision for 1000-s Run (%)	204-Rod PWR Fuel Assembly Uncertainty (2σ) (Rods)
Neutron generator	± 1.5	6.1
AmLi source	± 0.6	2.4

APPENDIX

LOS ALAMOS NATIONAL LABORATORY EXPERIENCE WITH THE SANDIA MA-165 PULSED NEUTRON GENERATOR SYSTEMS

We have been using the Sandia MA-165 pulsed neutron generator system⁹ for more than 2 yr. Sandia National Laboratories provided us with detailed electrical and mechanical drawings of the control circuitry and the cooling/high-voltage insulation assembly. The complete control circuit and all mechanical assemblies required for the system were fabricated at Los Alamos, using only the drawings provided by Sandia. As with any new and unfamiliar system, we had the usual number of start-up problems. However, these problems were minor, consisting mainly of spurious high-voltage-associated noise pulses that interfered with one of the feedback control circuits. This problem was eliminated through the use of simple electromagnetic screening and through rerouting of some of the internal wiring in the chassis.

Overall, the performance of the MA-165 system has been satisfactory. We have been using our original MA-165 system in our Differential Dieaway System^{10,11} continually for about 2 yr. We are now using our fourth sealed tube, with two of the first three sealed tubes having performed in accordance with Sandia Laboratories average-lifetime estimate of about 4×10^7 pulses, based on extensive bench testing. We obtained between $2-3 \times 10^7$ pulses, with each tube being in service 6 - 9 months before failure. These were Controlatron sealed (D,T) tubes. An updated version known as the Zetatron is the current Sandia model. Our one premature failure occurred with our first Zetatron tube and, according to the Sandia personnel who checked out the failure thoroughly at our request, the failure was caused by either (1) an originally defective tube or (2) a faulty feedback circuit in our control unit, a problem that has since been rectified. We obtained about $2-3 \times 10^6$ pulses from this tube in about 3 wk of service.

The pulse-to-pulse uniformity from the generators is very good. We measure the neutron output routinely and have found no degradation over either a short-term radiation period or over a period of several months of hard, continual use. The Sandia operating manual⁹ discusses the proper warm-up, the initial fine tuning of the tube high voltage and the emitter pressure feedback control parameters, and the general "folklore" for long lifetime of the (D,T)

tube. We recommend discussions with either L. G. Rice or G. W. Smith of Sandia National Laboratories, Albuquerque, for anyone contemplating use of this system.

The nominal output characteristics of the MA-165 system are 10-15- μ s duration pulses of 10^6 14-MeV neutrons each and repetition rates variable from 1-100 Hz. The system is operable with standard (US) 110-V ac electrical power, and the (D,T) tube is easily replaced. The unit has dimensions of 3.8-cm diam by 10.8 cm long and contains about 10 Ci of tritium. No tubes have been known to leak tritium.

Our estimate of cost for the MA-165 system is about \$30K. This estimate includes all parts and labor charges for fabricating the required control circuits and tube housings. Replacement tubes can be purchased from Kaman Sciences, Inc., of Colorado Springs, Colorado, for about \$6300 each.

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