Online Analyzers: Cf-252 Supply Chain Update and Risk Mitigation

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Abstract

Several hundred cement producers use online bulk material analyzers to help improve their quality control and plant efficiency. The predominant technology used for online analysis is Prompt Gamma Neutron Activation Analysis which requires a source of neutrons to function. This paper reviews the long-term availability of neutrons from the isotope californium 252 (Cf-252) as well as from an alternate source of neutrons produced electrically from a small linear accelerator otherwise known as a neutron generator.

Introduction - Neutrons for Online Analysis in the Cement Industry

More than 500 cement plants around the globe rely on online elemental analyzers to help control both raw material and kiln feed quality and reduce their variability. These analyzers use Prompt Gamma Neutron Activation Analysis (PGNAA) and in some cases Pulsed Fast Thermal Neutron Analysis (PFTNA). Both techniques require that the material being analyzed be activated by neutrons. The neutrons interact with, and briefly activate, the raw materials on a conveyor to subsequently generate gamma rays. The gamma rays created are counted and their energies analyzed to determine the elemental composition of the raw materials.

There are two sources of neutrons for online bulk material analysis: 1) the radioisotope californium 252 (Cf-252) and 2) neutron generators. Currently, there are four companies manufacturing online analyzers for the cement industry, all of them using neutron interrogation techniques. Two of these companies manufacture Cf-252-based systems only, one manufactures neutron generator-based systems only, and one company manufactures both types of systems.

Cf-252 and Supply Chain Risk Mitigation

The predominant source for neutrons enabling online analysis is the radioisotope Cf-252. More than 90% of all online analyzers installed today use Cf-252 as a source of neutrons. Cf-252 is a radioactive isotope that is man made in special high flux nuclear reactors. Cf-252 fissions spontaneously and continuously, emitting neutrons over a spectrum of energies. The energy spectrum has an average of 2.1 MeV and a most probable energy of 0.7 MeV (Figure 1). Cf-252 is unique as a neutron source in that it is compact and extremely reliable. The half-life of Cf-252 is 2.6 years which leads to a requirement for periodic “replenishment” over the life of an online instrument.

Cf-252 is produced by irradiating curium-244 (Cm-244) targets in these special reactors. Through a nuclear transmutation process of neutron absorption and beta decay, a significant fraction of the Cm target becomes Cf-252. After removal from the reactor, the target is chemically processed to remove the Cf-252 which is then plated onto a palladium alloy wire. An appropriate amount of this wire, dependent on the neutron activity desired, is cut and then doubly encapsulated into small zirconium alloy or stainless steel containers (Figure 2). PGNAA online analyzers use these small capsules.

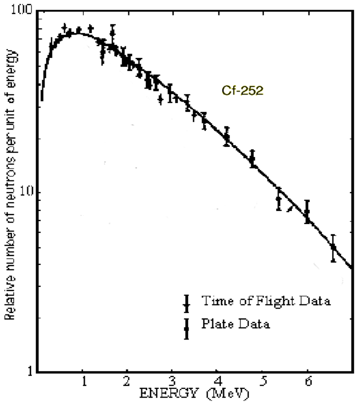

Figure 1 – Cf-252 energy spectrum

Figure 2 – Cf-252 capsules

There are multiple industrial uses of Cf-252. In addition to online bulk material analysis, Cf-252 is used to start nuclear reactors, for nuclear reactor fuel rod scanning, for oil exploration (via down-hole well logging), for the medical treatment of cancer, for off-line materials analysis, for materials inspection, and for research.

There are two High Flux Isotope Reactors (HFIR) in the world which can be used to create Cf-252. One is in the United States and one is in Russia, with the former accounting for the majority of the production. The reactor in the United States is located in Tennessee at Oak Ridge National Laboratories (ORNL) and is controlled by the United States Department of Energy (DOE). This reactor produces approximately 2/3 of the global supply of Cf-252 while the remaining 1/3 comes from the reactor at the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad, Russia (Figure 3).

Figure 3 – Oak Ridge National Laboratories Facility in Tennessee, U.S.A and Research Institute of Atomic Reactors in Dimitrovgrad, Russia

The production capacity of these two reactors to manufacture Cf-252 significantly exceeds not only current global demand, but predicted future needs as well. The current total demand for Cf-252 is estimated to be 30–40 mg/year and of this, the annual need for Cf-252 for online PGNAA systems is about one third of that total. As such, PGNAA analyzers represent the largest single application of Cf-252 in the world. With this in mind, there was more than a little concern among analyzer manufacturers and cement producers, when, in May 2008, the DOE announced without warning that it planned to halt production of Cf-252 in the ORNL reactor beyond the end of 2008.

A year and a half after this initial announcement, a very different picture has emerged. Not only has the DOE retracted its initial announcement and has *not* halted production of Cf-252, it has actually increased its output of this man-made isotope. The reversal in the DOE position on Cf-252 was achieved because of an aggressive and prompt response by industry to this potential disruption in supply. The swift reaction on the part of industry included meetings with the DOE and Congressional representatives.

During discussions with the DOE, industry representatives learned that the precipitous announcement had arisen because the US government's need for Cf-252 had gone away, and the DOE had merely assumed that private industry would not be willing to make up the resultant funding gap to continue the Cf-252 program. However, by the end of August 2008 common ground was established and a path to a successful resolution was visualized. It took another nine months to work out the details of an agreement, but in May 2009, the DOE, the industrial users, and the source encapsulators all came together in a legal agreement. That document assures the supply of Cf-252 from the DOE through 2012, with every expectation of continued supply beyond that year. At this time discussions continue between private industry and the DOE to address availability and pricing of Cf-252 when the current 3-year agreement expires.

While the primary purpose of the ORNL HFIR reactor is high technology research, the operating schedule of the reactor readily accommodates the production of Cf-252. Suggestions by industry representatives in August 2008 led to a change in the Cf-252 production process which increased efficiency and cycle capacity by 50% without any modification to hardware or need to add personnel. As well, the HFIR reactor and the Cf-252 recovery infrastructure have recently received substantial federal funds to put the program on track for an extended life, with the reactor itself anticipating another 40 years of use.

The cement industry can be proud of the role played by the US analyzer manufacturers in the campaign to overturn the DOE May 2008 decision. They engaged in lobbying with members of Congress and participated in all the major meetings between the DOE and private industry. Both are signatories to the DOE agreement and have stepped in to address the funding gap which the DOE incurred, once the US government demand for Cf-252 went away.

With the Cf-252 production capacity not a limiting factor and the long-term supply chain of Cf-252 assured, users of isotope-based online analyzers will continue to benefit from the process optimization and cost savings these unique instruments provide for a long time to come. Those interested in employing these systems are able to take advantage of the reliability and cost effectiveness of Cf-252 as a source of neutrons.

Neutron Generators - an Option for Online Analysis

While Cf-252 provides a reliable, cost-effective source of neutrons for online analysis, an alternate source of neutrons exists. Neutrons can be generated electrically through the use of a compact linear accelerator, also known as a neutron generator. These devices induce a fusion reaction by accelerating either deuterium atoms, tritium atoms or a mixture of these two isotopes into a special target also containing deuterium, tritium or a mixture. The result of the fusion reaction is an isotope of helium and a neutron.

The main component of a neutron generator is its linear accelerator, commonly known as the neutron tube. The neutron tube consists of an ion source (anode, cathode, magnet, pulser and gaseous deuterium and/or tritium), ion accelerator, focusing components and a target. Ions created in the ion source are accelerated and focused onto the target (Figure 4). When the ions strike the target with high energy, the resulting reaction liberates a neutron and an isotope of He.

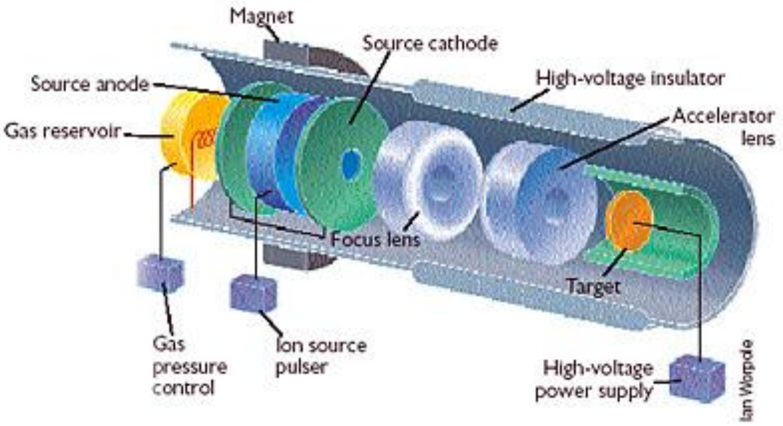

Figure 4 – Neutron Generator Tube

The neutron tube needs high voltage and control in order to operate. Therefore, separate power supply and control devices are used in conjunction with the neutron tube. A neutron tube coupled with components of the high voltage power supply is called the accelerator head and typically weighs on the order of 12 Kg (25 lbs) and may have approximate dimensions of 60 cm in length X 13 cm in diameter (24” length X 5” dia.) (Figure 5).


[redacted]

Figure 5 – Neutron Generator Accelerator Head with Control Components

Neutron generators used in online analyzers use deuterium and tritium and emit neutrons of 14 MeV, much higher than the mean energy of Cf-252 neutrons. Unlike Cf-252, neutron tubes require replacement periodically due to failure. The life of the neutron tube is variable with a relatively wide distribution that is highly correlated to the level of neutron output and operating conditions. A low neutron output will provide, in general, a longer average tube life and conversely a high neutron output will mean, again in general, a shorter average tube life. Neutron tube degradation and expected lifetime can be monitored, to a degree, by monitoring the high voltage required to maintain a constant neutron flux. As the neutron tube degrades, higher voltage must be applied to maintain a constant flux. However, exact time of failure cannot be predicted.

Because of this relationship between neutron output and tube life, the most commonly found application of neutron-generator based analyzers is stockpile control. That outcome derives from the fact that stockpile analyzers may operate 40-50 hours per week, while quarried material is being moved, while raw mix analyzers operate 168 hours per week. With the 24/7 duty cycle of raw mix analyzers coupled with the need for higher precision (and hence higher neutron output) in that application, the expected tube life can be as much as 5-6 times shorter than that found in stockpile applications. Many cement producers would find the ongoing costs associated with tube replacement in the raw mix applications problematic.

Online analyzer repeatability performance is correlated to neutron flux; the higher the neutron generator output, the better the repeatability performance. When evaluating an online analyzer application using a neutron generator, average tube life and expected replacement frequency should be taken into account. In essence, turning up the output of the generator to improve repeatability performance is at the expense of tube life (Figure 6).

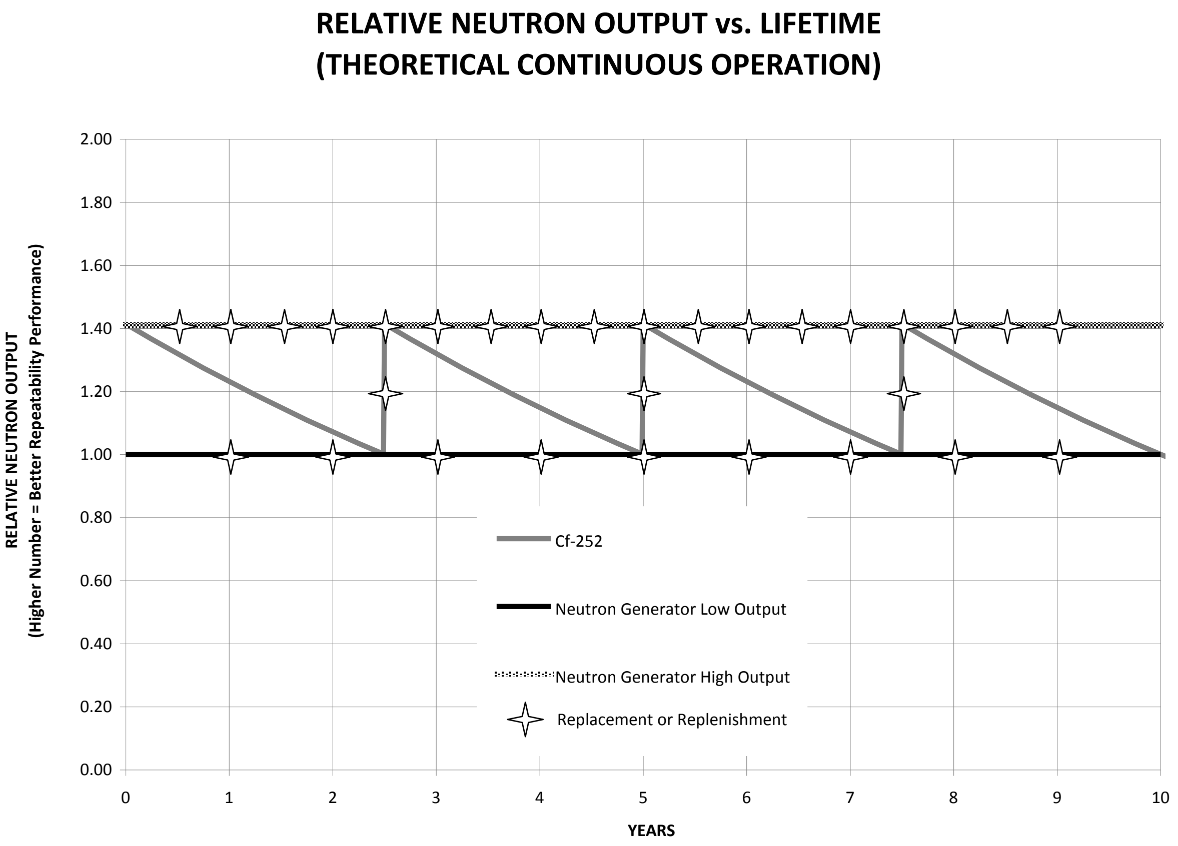

Figure 6 – Cf-252 and Neutron Generator Replacement Frequency vs Output

Cf-252 and Neutron Generators: a Comparative View

Cf-252 has been the preferred choice for neutrons among most online analyzer owners for various reasons. As mentioned earlier, chief among these are lower cost, reliability, and the simplicity of the isotope solution. For example, power, electronics and control are not required, and hence have no associated risk of failure. The lower cost applies not only to the source itself, but to shielding cost as well, since neutrons from generators have a higher energy level, necessitating either more shielding, or the construction of an exclusion zone with fencing and interlocks.

On the other hand, neutron generators offer different advantages. The most frequently mentioned among these is safety, since generators can be turned off when the instrument is not in use. However, this safety advantage is offset to a degree by the increased energy level of the neutrons produced by these systems. The higher energy neutrons penetrate further, and while the generator is operational are capable of more biological damage than neutrons from Cf-252.

Another related advantage of neutron generators is ease of transport. Transportation arrangements, regulations, and documentation are all simpler with the neutron generator option.

There are two other advantages of neutron generators related to performance. One is the ability to maintain a constant neutron flux over the life of the product, whereas Cf-252-based analyzers experience a decrease in flux over the 2 ½ year period between replenishments owing to source decay. (However, a subtle point of note here is that in order to keep neutron generator tube replacement frequency within acceptable limits, the neutron output of generator-based systems may be kept relatively low as compared to the initial output of a similar Cf-252 based systems. In other words, a generator-based system may have lower neutron output than the output of a Cf-252-based system, even after 2 ½ years of decay.) A final advantage of neutron generators is the ability to analyze certain elements, such as oxygen and carbon, when operating the generator in a pulsed mode. For the cement producer, however, this feature may be of minimal benefit.

One additional difference between Cf-252-based systems and neutron generator-based systems relates to the positional capability of the neutron source. Being small, Cf-252 capsules have the ability to be placed in multiple positions within the analyzer system whereas, due to cost, a neutron generator tube is generally located in a single location in the center of the system. The location of the neutron source is important as it can affect the uniform measurement sensitivity throughout the cross-section of material on the conveyor, as it passes through the analyzer. An analyzer with a single neutron point source unavoidably measures the material near the centerline of the conveyor more than it measures the material on the sides. Analyzers with two sources spaced to the left and right of center have better cross-sectional uniform measurement sensitivity at the same given detector placement geometry (Figure 7).

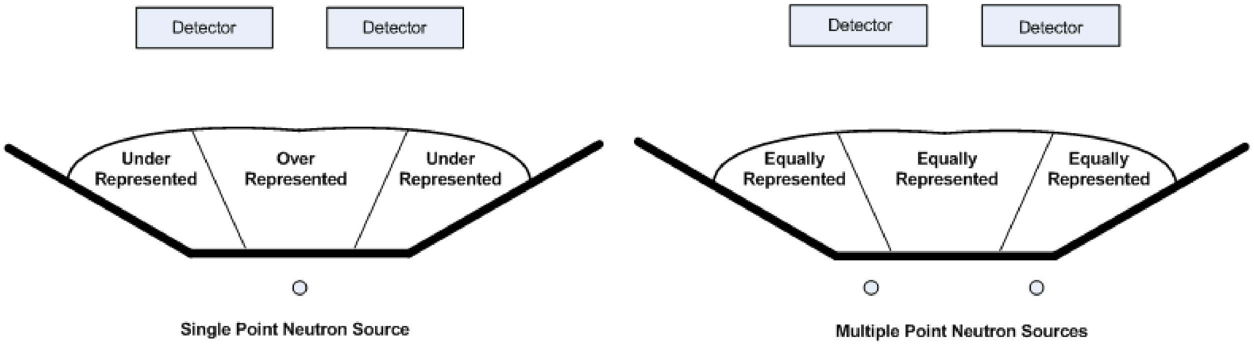

Figure 7 – Uniform Measurement Sensitivity Concept

Conclusion

In 2008 a threat to the long-term availability of Cf-252, the primary source of neutrons for online analyzers in cement plants, arose. That threat has been eliminated, with the recent decision by the DOE to continue production of this key isotope for the foreseeable future. In the meanwhile, there is a growing awareness of a second viable means of producing neutrons for online analysis, neutron generators. That option is available from more than one vendor, and while it suffers a cost and reliability disadvantage, it does offer some potential advantages. Either way, the long-term availability of neutrons for use in online analyzers for the cement industry is now on solid footing.

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