Available online at www.sciencedirect.com
ScienceDirect
Nuclear Instruments and Methods in Physics Research A 572 (2007) 1102–1105
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

# Candidate reactions for mercury detection induced by neutron and alpha particles

James J. Toth[a,*], Richard Wittman[a], Robert E. Schenter[a], John A. Cooper[b]

[a]*Pacific Northwest National Laboratory, 602 Battelle Blvd, Richland, WA 99352, USA*
[b]*Cooper Environmental Services, Portland, OR 97223, USA*

Received 10 October 2006; received in revised form 15 December 2006; accepted 19 December 2006
Available online 3 January 2007

**Abstract**

This paper summarizes modeling of mercury to activated states with alpha particles, neutrons, or deuterons, and the spectral emission from the activated products. Activation can occur with a neutron source such as $^{242}$Cf, $^{241}$Am-Be, a neutron generator, or a particle accelerator, and the activation products are measured if sufficient signal is provided. Identification and measurement of mercury by prompt gamma emission, generated by bombardment with neutrons are reported. Activation product reactions of ($\alpha$, $x$n) (d, $x$n) (n, $\gamma$) and (n, p) are screened as candidate reactions. In addition to prompt gamma emission, products and cross-sections of delayed activation involving alphas, deuterons and neutrons are also reported.

Initial calculations indicate the potential use of either alpha, or 14 MeV neutron activation to assess part per billion concentrations of mercury in the gaseous phase. Ultimately, data from sample analysis of ambient-condition flue gas will be used to assess mercury detection sensitivity and specificity under typical operating conditions.


*Keywords:* Fast neutrons; Modeling; Prompt Gamma Neutron Analysis (PGNAA); Mercury; Isotopic composition

## 1. Introduction

The importance of mercury and methyl mercury contamination in the environment and to human health is underscored by the fact that the US Centers for Disease Control tested the blood mercury levels of a representative sample of women of childbearing age. Based on these CDC data and new research, more than 300,000 newborns each year in the United States may be exposed to mercury concentrations higher than those considered to be without increased risk, i.e. at or above 5.8 μg/L [1]. In utero exposure of mercury leads to increased risk of adverse neurological effects. The National Research Council recommended a reference dose of 5.8 μg/L. Exposure of mercury leads to increased risk for adverse neurological effects [2].

The current levels of mercury emitted from the 1,300 coal-fired utility boilers in the US represent the greatest contribution of mercury release to the environment. On March 15, 2005, the EPA issued the first-ever federal rule to permanently cap and reduce mercury emissions from coal-fired power plants. When fully implemented, the clean air rules will reduce utility emissions of mercury in the United States from 48 tons a year to 15 tons, a reduction of nearly 70%.

No widely used technologies have been developed that can accurately measure total mercury below about 5 μg/dry standard cubic meter of flue gas. The current technology is based upon cold-vapor atomic adsorption analysis that represents total mercury in a stream of gas. An example analyzer is available from Nippon Instruments Corp.[1]

This paper relates to the modeling of mercury activation or decay reactions and an experiment involving a neutron or particle generator, producing a neutron or charged-particle fluence, and a detector to measure total mercury in

*Corresponding author. Tel.: +1 509 376 1723; fax: +1 509 376 9781.
*E-mail address:* james.toth@pnl.gov (J.J. Toth).

[1]Environmental Technology Verification Program, US Environmental Protection Agency, Continuous Emission Monitor, Nippon Instruments DM-6D/DM-6P.

0168-9002/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2006.12.032

Table 1
Mercury (Hg) stable isotopes

| Isotope | Percent natural abundance |
|---|---|
| $^{196}$Hg | 0.15 |
| $^{197}$Hg | 0.00 |
| $^{198}$Hg | 9.97 |
| $^{199}$Hg | 16.87 |
| $^{200}$Hg | 23.10 |
| $^{201}$Hg | 13.10 |
| $^{202}$Hg | 29.86 |
| $^{203}$Hg | 0.00 |
| $^{204}$Hg | 6.87 |
| Total | 100.00 |

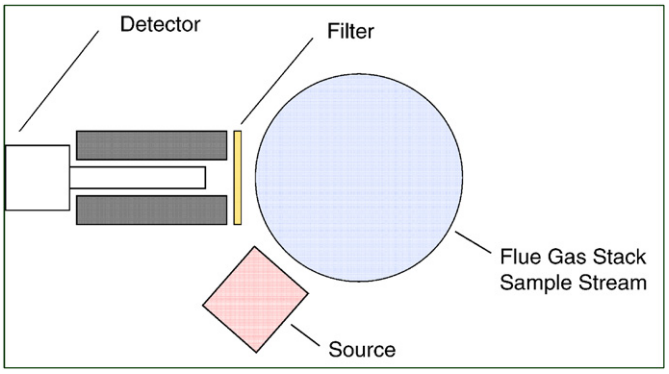


Fig. 1. Schematic arrangement of new mercury detector using prompt gamma activation analysis.

gases. The natural abundance of mercury is provided in Table 1.

A slip stream of flue gas exiting from the precipitator is routed to the instrument facility. The required volume of flue gas required for interrogation is up to 1 L. High-purity germanium detectors count fractions of reaction events. For delayed activation analysis (item 2 below), a well-type high-purity germanium detector would be used, where the activation sample passes through the middle of the detector. Detector efficiencies on the order of 10–30% are expected.

In principle, with respect to the time of measurement, activation analysis falls into two categories:

(1) Prompt Gamma Neutron Activation Analysis (PGNAA), where activation measurements take place during irradiation.
(2) Delayed Activation Analysis where measurements follow the radioactive decay.

Calculations in the following two sections estimate activities for reaction products for case (1) PGNAA and the delayed activation products.

## 2. Prompt neutron activation analysis

The concept of prompt neutron activation analysis of mercury in flue gas is shown in Fig. 1. In this case, the PGNAA method depends upon a high neutron flux to activate and measure the gamma emission with a sufficient intensity [3]. The energy of the gamma ray is indicative of the isotope present. Note below is provided by Bell, 1994 [4]:

> The thermal neutron cross section of mercury is 384 barns and is due almost entirely to the 2200 barn cross section of the isotope Hg-199. However, the resultant Hg-200 is stable, and therefore cannot be detected by delayed activation analysis. On the other hand, 77 percent of the de-excitations of the Hg-200 involve the emissions of a 368 KeV gamma ray. The gamma may be detected by commercially available germanium detectors.

Given a 14 MeV neutron source, a surrounding hydride (or deuteride) blanket can supply a thermalized neutron source to the sampe volume. For a thermal flux of $1 \times 10^9$ neutrons cm$^{-2}$ s$^{-1}$, the resulting gamma activity for $N_{\mathrm{Hg}}$ mercury atoms is

$$R_\gamma = \phi_{\mathrm{n}} \sigma_\gamma f_{199} N_{\mathrm{Hg}} r_{368\,\mathrm{keV}} \quad (1)$$

where $\phi_{\mathrm{n}}$ is the neutron flux $= 1 \times 10^9\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ (selected basis), $\sigma_\gamma$ the neutron capture cross-section $= 2200 \times 10^{-24}\,\mathrm{cm}^2$, $N_{\mathrm{Hg}}$ the mercury atoms in sample (natural isotopic fractions), and $f_{199}$ the atomic fraction of $^{199}\mathrm{Hg} = 0.1687$, $r_{368\,\mathrm{keV}}$ the activation fraction for $^{200}\mathrm{Hg}(368\,\mathrm{keV}) = 0.77$.

The gamma emission at 368 keV for 1 mg (0.001 g) is calculated to be

$$R_\gamma = (1 \times 10^9\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1})(2200 \times 10^{-24}\,\mathrm{cm}^2)(0.1687) \times \frac{0.001\,\mathrm{g}}{200\,\mathrm{g\,mol}^{-1}} \frac{6.022 \times 10^{23}}{1\,\mathrm{mol}} (0.77).$$

Total activity: $R_\gamma = 8.6 \times 10^5\,\mathrm{s}^{-1}$ for the 368 keV emission rate of $^{200}$Hg.

Total activity $= 8.6 \times 10^5\,\mathrm{dis\,s}^{-1}$ or about 23 μCi.

The basis of the calculation indicates a vapor sample containing 1 mg mercury will produce an activation of $5\mathrm{E}+6\,\mathrm{dis\,s}^{-1}$, given a neutron flux of $1\mathrm{E}+09$ neutron cm$^{-2}$ s. If the basis of the calculation is replaced with 1 ng of mercury, the total produced activity becomes 23 pCi. The expected detection limit will depend upon background [5] and count length of time, and is expected to be sub-μg quantity of mercury.

## 3. Delayed activation analysis of radioactive decay products

Delayed Gamma Neutron Activation Analysis (DGNAA) may be useful for the detection of the radioactive nuclides that are produced from the stable isotopes of mercury. The sensitivity of the long-lived radionuclides suffers from the interference of the shorter radionuclides,