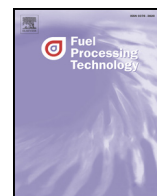




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Research article

A portable, continuous system for mercury speciation in flue gas and process gases

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ABSTRACT

During fuel combustion, mercury in the form of Hg^0 and Hg^{2+} is emitted to the atmosphere. Effective reduction of mercury emission requires the application of speciation systems for emission control and research. To study the speciation of mercury in flue gas a portable and continuous system was constructed. The speciation system is based on the wet method of Hg^{2+} transformation to Hg^0 . The main part of the system is the Nippon Instrument Corporation EMP-2 WLE-8 set. In order to operate in flue gas from coal burning the Nippon Instrument Corporation set was equipped with the following devices: fly ash filter, steel probe, transfer line, and the tee connector. The portable, continuous system was successfully tested in the laboratory and during the industrial tests in Poland and the Czech Republic. The described system was also used to determine mercury in the process gases (low-temperature pyrolysis of coal). The results of mercury speciation in flue gas were compared with Durag HM 1400 TRX system.

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1. Introduction

The combustion of solid fuels is one of the main sources of environmental contamination with mercury compounds [1–6]. Hg in coals exists in the forms of several compounds [7,8]. Mercury is present within untreated coal-derived flue gases at trace levels of around 1 ppb. At the high temperatures within the boiler, the mercury is liberated from the coal as elemental mercury. As heat is extracted from the combustion process, mercury can exist within the flue gas as elemental mercury, oxidized mercury (compound form, often assumed to be mercuric chloride), and particulate-bound mercury [9–13]. Unlike oxidized mercury, Hg^0 is insoluble in water and may be transported for long distances. In the form of Hg^{2+} mercury may be bound to airborne particles [14] and can be introduced into the body by inhalation. Hg^{2+} may also reach the water reservoirs with rain. In water mercury can be transformed to toxic organic forms such as MeHg, which are a threat to living species [15,16]. The low concentration makes distinguishing between the three forms very difficult. What complicates the analysis further is the fact that mercury can readily convert between these three species, and because of the low concentrations, also be readily lost due to adsorption on tube walls [9–13]. Moreover, various surfaces such as tube walls can catalyse the oxidation of mercury [11,12,17].

Toxicity of mercury caused the adoption of a series of legal regulations. In the USA in 1990, the Clean Air Act put the mercury on the list of 189 particularly Hazardous Environmental Pollutants (HAPs). In

2005, the Environmental Protection Agency (US EPA) ordered the reduction of mercury emissions from fuel combustion processes in the US by approx. 70% by 2018. In 2011, the EPA introduced new standards for mercury emissions amounting to an average of $2.5 \mu\text{g Hg}/\text{m}^3_{\text{N}}$ for newly established power plants and $8.5 \mu\text{g Hg}/\text{m}^3_{\text{N}}$ for existing plants [18]. The European Community also introduced regulations aimed at reducing mercury emissions (Integrated Pollution Prevention and Control Directive 96/61/WE, Large Combustion Plant Directive - 2001/80/WE, National Emission Ceilings Directive - 2001/81/WE). These regulations stipulate regular monitoring of pollution generated from fuel combustion installations with a nominal power > 50 MW, the reporting of results through the European Pollutant Release and Transfer Register (E-PRTR) and the use of the Best Available Technique (BAT) that limit emissions. Moreover, there is the Industrial Emissions Directive (IED) of the European Parliament (2010/75/EU of 24.11.2010) on industrial emissions. From 1 January 2016 a requirement is imposed to carry out at least a single measurement of total mercury per year (for coal-fired emissions of the sources of power > 100 MW).

On the market today there is a predominance of stationary, continuous systems for mercury speciation such as: Sir Galahad II (PS Analytical), Tekran 3300 or the Mercury Freedom System (Thermo Scientific) [19–22]. To measure mercury a few times per year the small, portable analytical systems are economically more cost effective than stationary systems working continuously all year round. Good examples of portable systems for mercury speciation in flue gas are the Ohio Lumex RA-915+ with the sorbent Trap Sampling System OLM30B and the RP-M324 Attachment for Sorbent Trap Analysis [23]. The United States Environmental Protection Agency has built, on the basis of the

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Lumex trap method, the Mercury Measurement Toolkit. The Toolkit was successfully used to develop emission factors for coal-fired power plants in such countries as China, Russia and South Africa [24]. For many years the reference method for mercury speciation in flue gas was the Ontario Hydro Method [25–27]. However, in the recent years its popularity has diminished rapidly. Its basic disadvantage is the inability to speciate mercury continuously, a high consumption of reagents and difficulty of use in industrial environments.

The purpose of this article is to provide the reader with information on constructing, tests and use of the small, portable and continuous system for mercury speciation in the flue gas and process gases (PC-AGH). The PC-AGH system is based on the wet method of Hg^{2+} to Hg^0 transformation. The main part of the system which is presented is the Nippon Instrument Corporation EMP-2 WLE-8 set. In order to operate in flue gas from coal burning the Nippon set was equipped with the following devices: fly ash filter, a steel probe, a transfer line and a tee connector. The article is based on three years of experience in the construction, testing, modification and industrial use of our continuous, portable system for mercury speciation in flue gas.

2. Materials and methods

2.1. Produced apparatus

The EMP-2 WLE-8 mercury speciation set made by Nippon Instruments Corporation (NIC) was used as the basis for the portable continuous system for mercury speciation (PC-AGH) in flue gas (Fig. 1). The PC-AGH system was designed to study the speciation of mercury in the flue gas as well as to observe the dynamic changes of mercury concentration in processes such as low-temperature pyrolysis or tests of sorbent performance. The principle of operation of the NIC speciation set is a continuous, simultaneous measurement of total mercury and mercury at zero oxidation state. The NIC speciation set uses a wet method (SnCl_2 solution) for Hg^{2+} reduction. Concentration of Hg^{2+} in gas is calculated from the difference of total mercury concentration and concentration of mercury at zero oxidation state. The measurement range of the EMP-2 detector is from 0.1 to 999.9 $\mu\text{g}/\text{m}^3$. The response time of the detector is 1 s.

The NIC speciation set consists of two EMP-2 AAS (Fig. 1-1) mercury analysers and two scrubber units (Fig. 1-2). Each scrubber unit consists of three scrubbers. The first scrubber in the first unit is filled with 10% SnCl_2 which reduces Hg^{2+} to Hg^0 . According to our own test and the NIC test, the applied wet reduction method provides 100% transformation of Hg^{2+} to Hg^0 [28]. The first scrubber in the second unit is filled with 10% KCl which removes Hg^{2+} ions from the flue gas stream. The second scrubbers in both units are filled with a 10% KOH solution

which removes acid components of flue gas and protects detectors. The last scrubbers in both units are empty and reduce the humidity from flue gas. The scrubbers are cooled down to about 6 °C by means of the thermoelectric effect.

The EMP-2 detector works in two measurement modes: mobile and stable. In the mobile mode the resolution of the EMP-2 detector is 1 $\mu\text{g}/\text{m}^3_{\text{N}}$ and in stable modes 0.1 $\mu\text{g}/\text{m}^3_{\text{N}}$. In both modes the detector performs one measurement per second. According to the NIC information, the EMP-2 WLE-8 basic set is ready for speciation of mercury in the process gas that does not contain solids particles such as fly ash. In order to operate in flue gas from coal burning and process gases the set was equipped with the following devices: fly ash filter (a construction of our own) (Fig. 1-4), steel probe (ZAM Kęty, Poland) (Fig. 1-3), transfer line (JCT, Austria) (Fig. 1-5), and the tee connector (a construction of our own) (Fig. 1-6). The sampling system is consistent with the guidelines of the Ontario Hydro Method (ASTM D6784 - 02(2008)).

The system includes also the following additional devices constructed by ourselves: ash filter heater, Hg^0 generator (own construction) – used mainly for the preparation of calibration gas with a high Hg^0 concentration, Hg^{2+} ultrasonic calibrator (a construction of our own) [29] used for Hg^{2+} sorption check and system tests. Below one can find the descriptions of untypical elements of the PC-AGH system.

2.1.1. Fly ash filter

In the preliminary version of the mobile system the typical ceramic industrial filter (JCT, Austria) with the reverse air flow for filter cleaning was applied. The filter was placed after the probe and transfer line. Unfortunately, the industrial tests showed that after first use the Hg^0 sorption on the probe, transfer line and filter reached 90%. The filter cleaning using filtered compressed air did not result in a decrease of sorption. A decision was made to change the order of items in the measurement system and to use the filter of our own construction (Fig. 2). The filter is the first device in the sampling line. During the measurement the filter is heated by flue gas. Outside of the flue gas the tract filter is heated by an ash filter heater. The ash filter casing is made of stainless steel. The interior part is made of two cylindrical pieces of PTFE. As the filtration element 37 mm glass filters are used. Such a filter design facilitates quick replacement of the filtration element and also facilitates PTFE elements exchange in case of filter contamination. The filter was designed to perform measurements not only in the chimney, but also when the dust concentration is at the level of several dozen g/m^3 (e.g. before the electrostatic precipitator). Of course, in such conditions, the time of measurement is in the range of 10–20 min in contrast to the measurements on the chimney, where measurement may last a few hours.

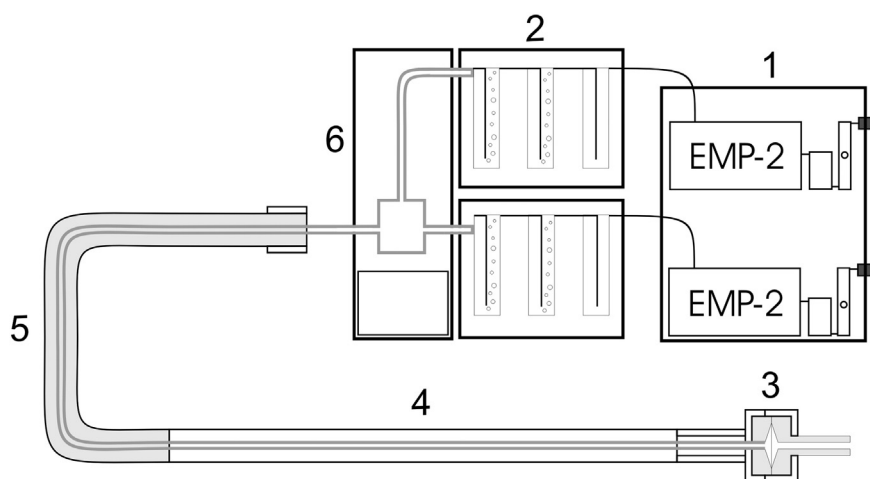


Fig. 1. The design of the mobile system for mercury speciation in flue gas.

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