

Determination of total mercury in coke oven gas

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ABSTRACT

Coke oven gas is one of the products of the coal coking process. Coke oven gas is used mainly as fuel in the coking plant. Additionally, the high density of hydrogen predisposes the coke oven gas to be used as an environment-friendly, low-CO₂ fuel or as a raw material in the chemical industry to produce methanol [4–8] or syngas. Due to the reasons mentioned above, the knowledge of coke oven gas pollutants such as mercury is important. The source of mercury in the technological installation of the coking plant is the coking of coal that may contain various compounds of mercury. The average concentration of mercury in charge coal measured in coals from four coking plants was 75±9µg/kg. During the heating of coal the concentration of mercury in coal is reduced. The average concentration of Hg in coke measured by Konieczynski was 14±7µg/kg (about 20% of Hg which exists in charge coals). This means that about 80% of mercury from coal enters various products such as sulfur, coal-tar and coke oven gas or is emitted in disorganized emissions during the loading of the coke oven battery [14]. Together with coke oven gas, mercury enters the technological circuit of the coking plant and then, with a flue gas, the environment.

Since 2007 all coking plants are subject to the European Pollutant Release and Transfer Register (E-PRTR). According to the legal regulations, coking plants are obliged to monitor the pollutants that are released and to issue reports. The threshold of mercury and its compounds released to the air, in accordance with the E-PRTR regulations, is 10 kg/year. The exceeding of this level obliges coking plants to inform the relevant authorities responsible for environmental pollution monitoring about this fact.

In the literature there is a lack of information on mercury determination in coke oven gas. Determination of mercury in the coal and solid or liquid products of the coking process such as: coke, desulfurization products or coal-tar is relatively simple and could be performed by means of a mercury analyzer. However, for the full balance, mercury should be also determined in the coke oven gas. The determination of mercury in process gases like flue gas is amply described in the literature, but from an analytical point of view, due to the rich organic matrix and hydrogen, coke oven gas is a much more complicated sample. The main components of coke oven gas are N₂, CO₂, CO, CH₄ and H₂, but even purified coke oven gas also contains a high amount of benzol (about 3g/m³_N) and naphthalene (about 0.5g/m³_N). For this reason, in the case of coke oven gas, the use of continuous analyzers or scrubber systems for the determination of mercury is impossible. In order to develop mercury balance in the coking plant, it was necessary to develop an effective and safe method for the determination of mercury in coke oven gas.

To enable the determination of mercury in the coke oven gas a sampling method (activated carbon traps) combined with a Cold Vapor Atomic Absorption Spectroscopy (CVAAS) determination of mercury was developed. As a part of the method optimization, the optimal flow rate (18 dm³_N/h) and sample volume (3 dm³_N) were determined. A significant effect of organic matrix on the quality of mercury determination in coke oven gas was recognized. The developed method of Hg determination could be used when the benzol concentration in coke oven gas does not exceed 9 g/m³_N. The method was validated according to the Eurachem recommendation. The assessed parameters were as follows: selectivity, limits of detection and quantification, working range, trueness and precision. The developed method was applied to mercury determination in the clean coke oven gas. The determined concentration of total mercury in the coke oven gas was 3.2±0.3 µg/m³_N (k=2) for n=18. The study showed that coke oven gas purification systems which exist in the coking plant can remove part of mercury from coke oven

gas. To the best of the authors' knowledge, this was the first instance of determination of total mercury in coke oven gas.