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Mercury emission and speciation from industrial gold production using roasting process

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ABSTRACT

Gold production has been identified as an important source of anthropogenic mercury emissions. Few measurements have been conducted on the mercury emission in the industrial gold production. In this study, field measurements on mercury concentration and speciation profile in the roasting flue gas were conducted in a two-stage roasting gold smelter, and the corresponding mercury emission factors were obtained using mass balance analysis. The average mercury concentration in the feed gold concentrates was 730.0 μg/kg, and the daily input mercury was 94.9 g in this research. In the roasting procedure, 38.4%, 27.3% and 9.0% of input mercury was removed into the sulfuric acid, contaminated water and arsenic, respectively, while 22.8% of input mercury flowed into the cyanidation-refining procedure within the roasting residue and the dust collected from the gas cooling tower and the electrostatic precipitator. Finally, 2.3% and 5.9% of input mercury was emitted into the atmosphere with the roasting flue gas and the refining flue gas. Mercury concentration in the roasting flue gas was 4.02 μg/m^3, where the proportion of Hg_p:Hg^{2+}:Hg^0 was 11:57:33. The balance calculation results indicated that the atmospheric mercury emission factor was 2.27×10^{-3} g mercury/g gold produced for the tested smelter. Both the emission factor and mercury removal efficiencies of air pollution control devices are useful for development of a more accurate emission inventory.

1. Introduction

Atmospheric mercury (Hg) has aroused public concern due to its health effects on humans and other organisms, and gold production has always being considered as a large source of anthropogenic Hg emissions (Lacerda, 1995, 1997). Based on the 2010 Hg emissions inventory, the amount of atmospheric Hg emission from gold mining, including artisanal and small-scale gold mining (ASGM) and large-scale gold production (LSGP, also known as industrial gold production), reached 824.3 t, taking the largest proportion (42.1%) of the global anthropogenic Hg emissions (AMAP/UNEP, 2013). Much of the historic emission of Hg was reported from ASGM, where Hg was employed for gold recovery from milled ore via amalgamation (AMAP/UNEP, 2013). This gold recovery method, however, has been effectively ceased in many countries due to its dramatic high Hg emission, and it has also been forbidden by law in China since 1997. Therefore, more attention should be paid on the Hg emission from LSGP in China in the future (Zhang et al., 2015).

In previous studies, great uncertainties existed in Hg emission factors (EFs) of LSGP. In general, there are four conclusive factors associated with Hg EF – the Hg concentration of feed gold concentrates, the Hg distribution factor to the flue gas, the type of air pollution control devices (APCDs) combination applied and the Hg removal efficiency of a certain type of APCDs combinations, and all of them are likely to vary considerably. Take the Hg concentration of gold concentrates for example, Fang et al. (2004) indicated that the gold concentrates generally contained 1000 μg/kg or more Hg by analyzing over 4000 gold concentrates samples in Taiwan, and the highest content was over 37,500 μg/kg. However, based on 43 concentrates samples in Shihu gold mine in China, the lowest Hg concentration was only 1.76 μg/kg, reported by Li (1990). Moreover, the Hg distribution factor to the flue gas is determined by the smelting technology employed. Obviously, compared with the heap leaching, the cyanidation with roasting pretreatment has more Hg release point due to thermal pretreatment process. Finally, based on the field experiments conducted in the non-ferrous metal smelting factories, it has been proved that the Hg of flue gas could be removed to certain extend with the utilization of the APCDs with different Hg removal efficiencies ranging from 2.4% to 99.2% (Zhang et al., 2012; Wu et al., 2012). Nevertheless, the aforementioned information was barely considered in previous researches. For example, in the research conducted by Argonne National Laboratory and Tsinghua University,

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0.79 g/g (g Hg emitted/g gold produced) was chosen as the LSGP Hg EF for lack of field measurements, and it was obvious that such Hg EF was overestimated since it was originally used to estimate the Hg emission from ASGM in China (Dai et al., 2003; Jiang, 2004; Streets et al., 2005; Wu et al., 2006). Pacyna et al. (2006) presented the LSGP Hg EF of 0.5 g/g in the Atmospheric Emission Inventory Guidebook (UN ECE, 2000) with no experiment specifications. UNEP estimated that the Hg EF of LSPG were 0.025–0.027 g/g and 0.055 g/g to calculate the Hg emission inventories of 2005 and 2010, respectively (AMAP/UNEP, 2008, 2013; Pacyna et al., 2010; Streets et al., 2011). However, only the distribution factor to air (0.04) and the Hg concentration of gold concentrates (55,000 μg/kg) were provided based on US national data, indicating that it is related to the specific technologies employed in the United States (AMAP/UNEP, 2013), and it failed to provide any specific information associated with the APCDs.

Since little information of field measurements on Hg emission of the industrial gold production is available, the main purpose of this work is to update the EF to the atmosphere in industrial gold smelter based on field measurements. In this paper, the concentration and speciation of Hg released from the roasting flue gas has been tested and the Hg removal efficiencies of the APCDs have been comprehensively discussed. Moreover, the behavior of Hg in the roasting flue gas and the fate of Hg through the whole smelting process have been systematically analyzed.

2. Experimental methods

2.1. Tested smelter

In China, the three technologies used in LSGP are heap leaching, roasting (including one-stage and two-stage roasting) cyanidation and bio-oxidation cyanidation, the latter two of which are utilized to deal with refractory gold ore. Compared with the heap leaching and bio-oxidation cyanidation, the roasting cyanidation has additional thermal pretreatment process, which is considered to be the largest Hg emission sources of LSGP (Miller and Jones, 2005). Besides, the processes in the heap leaching technology are included in the roasting cyanidation, and based on our investigation, bio-oxidation cyanidation is merely employed by one factory in Xinjiang province in China. Thus, in this research, a two-stage roasting smelter in Shandong province, which has the biggest gold production in China, was chosen for field measurements.

The process flow diagram of tested smelter is illustrated as Fig. 1. In this research, the whole smelting process is divided into two procedures – roasting and cyanidation-refining. The roasting procedure includes processes from the roasting to the flue gas emitting (processes illustrated in the green dot frame in Fig. 1);while the cyanidation-refining is the procedure during which the gold-bearing material, including the residue from the roasterII as well as the dust from U-shape gas coolerII (UGCII), gas cooling tower (GCT) and electrostatic precipitator (ESP), is cyanided to produce dore (processes illustrated in the blue dot frame in Fig. 1).

The operation temperature of the first and second roasting process is 550 °C for 1 h and 600 °C for half an hour, respectively. After the flue gas from two roasters passes through the corresponding UGCs, they merge together to go through the GCT and ESP successively. A small quantity of coarse particles collected by UGCI and the roasting residue of the roasterI go into the roasterII for further roasting. The temperature of the roasting flue gas at the inlet of the quench tower (QT) and the outlet of the fiber filter dust collector (FF) is 376 °C and 158 °C, respectively, so arsenic trioxide in the glue gas is condensed due to this dramatically cooling and then collected by FF. Recycled dilute sulfuric acid is utilized to clean the flue gas in venturi tube (VT) and packet tower (PT), and then electrostatic demister (ESD) is employed to remove the water vapor from the gas stream. The contaminated water and contaminated acid sludge is discharged after the roasting flue gas goes through VT, PT and ESD, and then the flue gas with high SO_2 concentration enters into the acid plant (AP). In the AP, the flue gas goes through the dehydration tower (DHT) and the double-conversion double-absorption (DCDA) process to produce H_2SO_4. In the cyanidation procedure, the gold-bearing material will dissolve in the NaCN solution to generate the pregnant solution with gold and other metallo-cyanide complexes (including Hg), in which the metals will be precipitated on zinc dust. Eventually, the zinc precipitate taken from the filter presses is refined at high temperature (over 1200 °C) to produce dore, during which all the available Hg will be released into the atmosphere since there is no APCD applied in tested smelter.

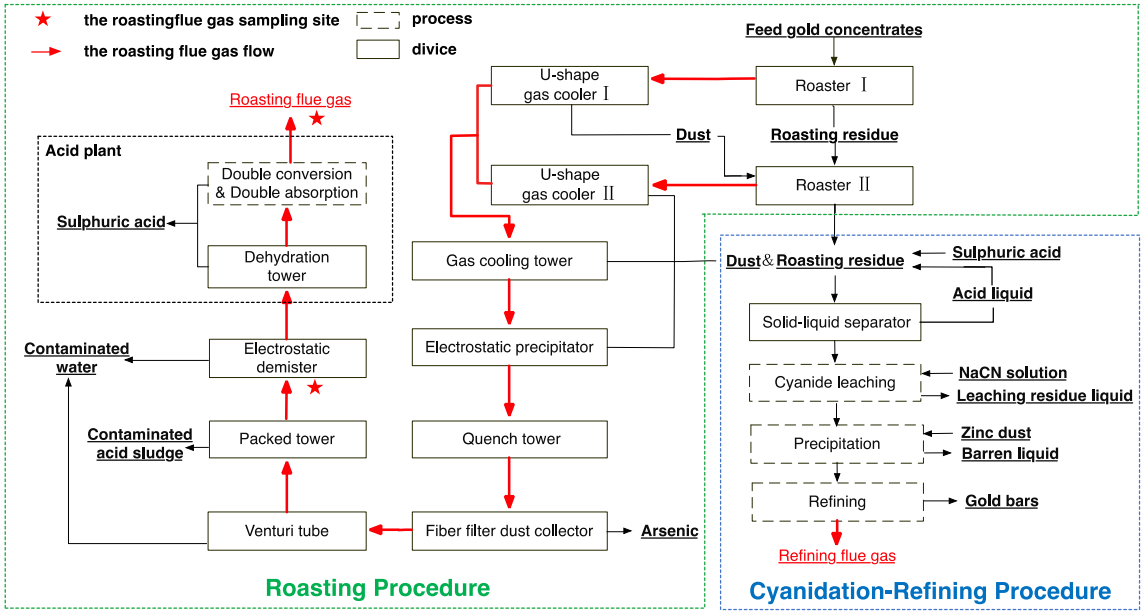

Fig. 1. Flow diagram in a two-stage roasting gold smelter.