Development of a novel mercury cartridge for mercury analysis

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Introduction

- Mercury cartridges are used to capture mercury from flue gases over a predetermined time interval and subsequently release concentrated pulses of mercury into a detection stream of high-purity carrier gas.

- Gold-coated silica beads(Au/SiO_2) are widely used in mercury cartridges for mercury preconcentration. The gold film on the silica beads forms an amalgam.

- When the mercury-loaded Au/SiO_2 is heated, a highly concentrated pulse of mercury is released and can be measured by a mercury analyzer.

Problems faced…

During mercury capture from a flue gas, the cartridge is transiently exposed to the acidic components of the flue gas.

Exposure to these components may interfere with cartridge performance by:

(i) affecting the efficiency of mercury capture through competitive Adsorption.

(ii) affecting mercury release by forming intermediate Species.

(iii) destroying the gold film, and releasing detrimental components into the mercury detector.

(iv) SOx, NOx and HCl are common components of flue gases and have broadband absorption at the detection wavelength of elemental mercury interfering with mercury detection by CVAFS and CVAAS.

In this study…

- A new material viz silver nanoparticles synthesized on the surface of natural chabazite(Ag/MC) was tried.

- It is synthesized by simple ion exchange followed by thermal annealing making it less-expensive alternative to the synthetic silica beads that are coated by gold through vapour deposition which require sophisticated devices.

In this study…

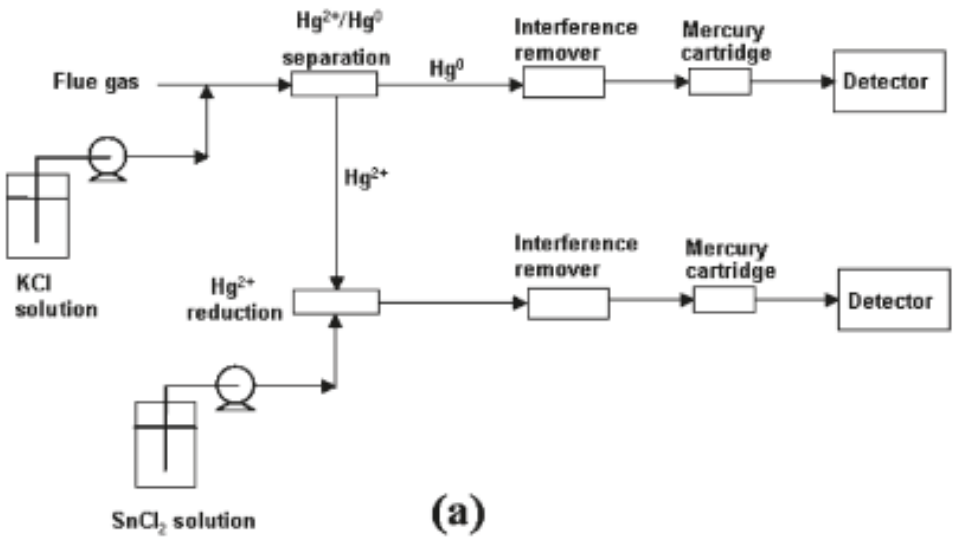
- A systematic investigation of mercury cartridge performance was conducted to demonstrate the critical role of mercury cartridges in mercury monitoring.

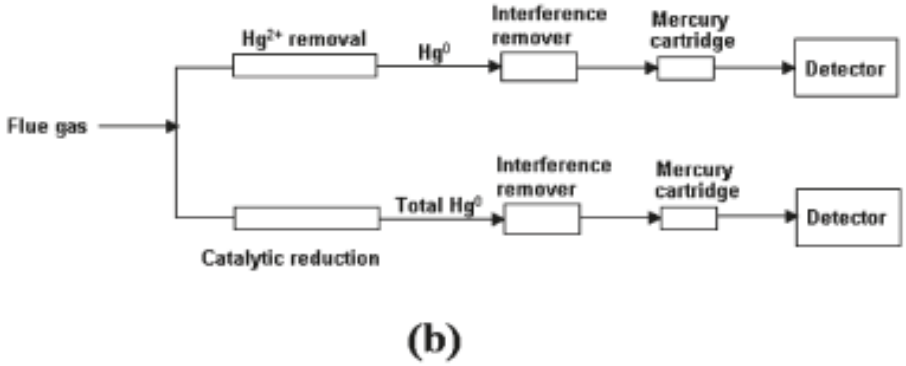
- Investigate the effects of two major interfering gas components(SO_2 and NO) on mercury cartridge performance.

- Two types of mercury cartridge sorbents were compared:

- Gold-coated silica beads (Au/SiO_2) and surface-supported silver nanoparticles formed on chabazite (Ag/MC).

- To mitigate the effects of SO_2 on mercury recovery from the Ag/MC mercury cartridge, a novel SO_2- and NO- tolerant mercury cartridge was designed and fabricated with soda lime placed upstream of the Ag/MC as a disposable SO_2 scrubber.





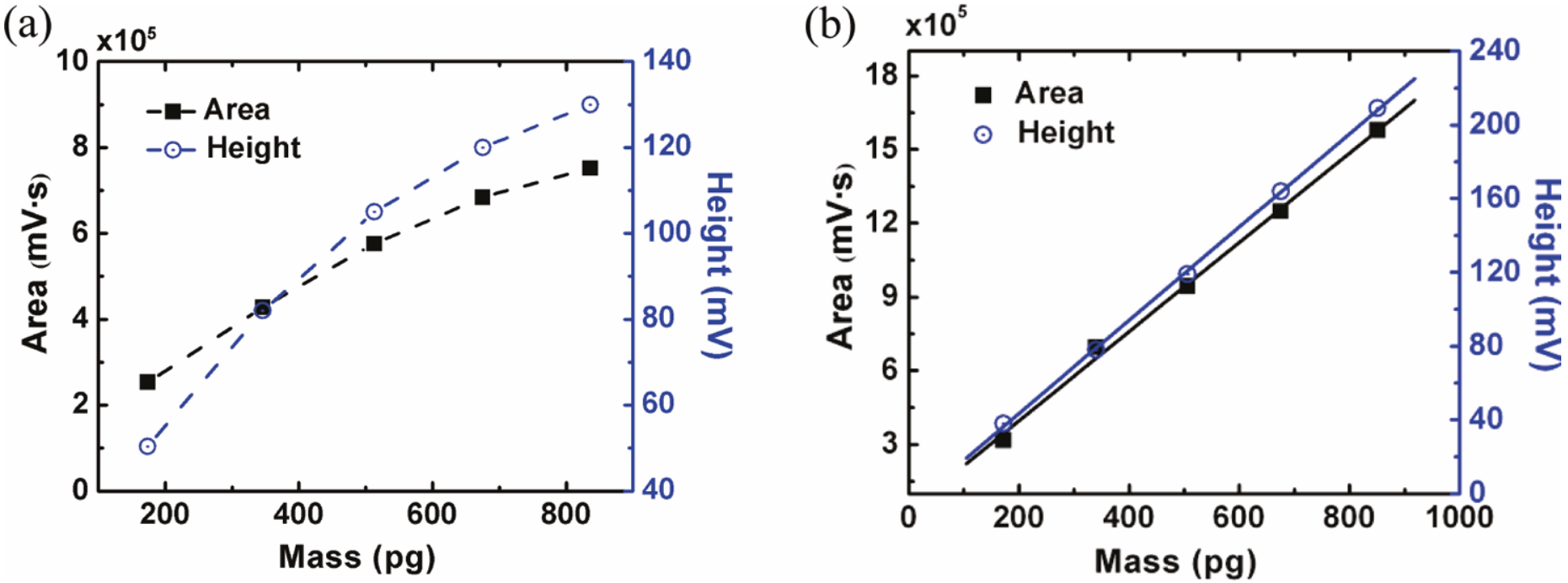
Principles of Semicontinuous Automated Mercury Monitors Utilizing (a) Wet and (b) Dry Speciation Approaches

Experimental

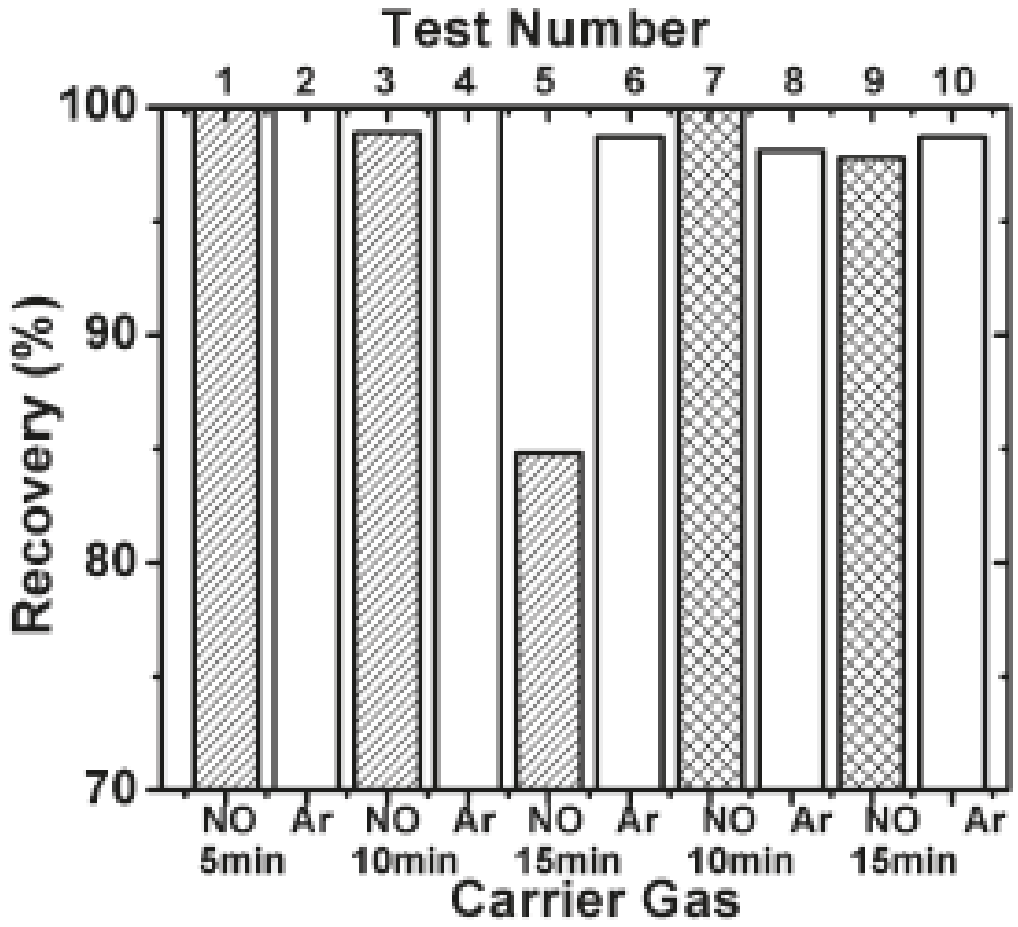
- Mercury cartridges consisted of a quartz tube packed with either Au/SiO_2 or Ag/MC and wrapped with a heating coil.

- Evaluation of Mercury Cartridge Performance in an Argon Stream. The experimental setup consisted of a mercury port, a mercury cartridge, and a mercury detector connected by Teflon tubing and fittings.

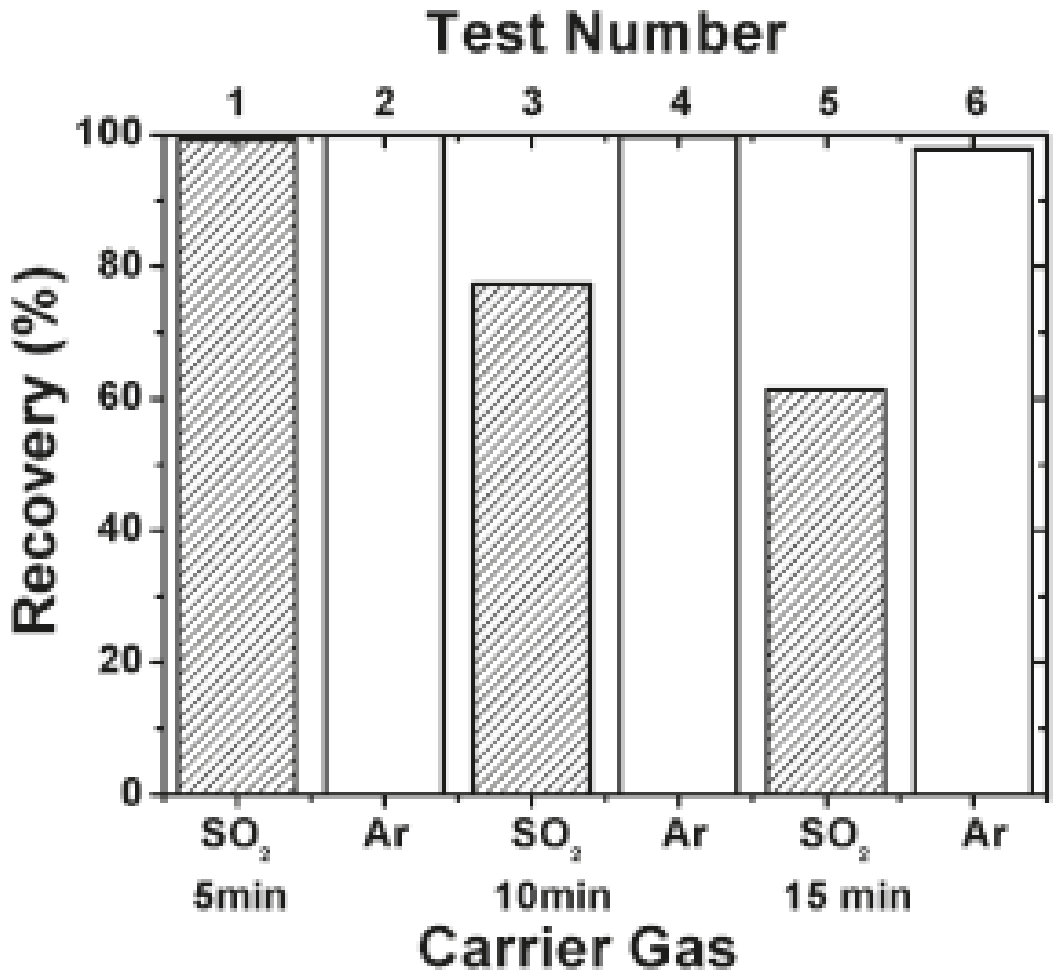
- A pulse of mercury-saturated air of known volume at storage temperature was injected into the argon stream from the mercury port. After 5 min, the mercury cartridge was heated by applying 5.6 V to the wrapped coil to release precaptured mercury. The released mercury was detected by a downstream mercury detector



Correlation of injected mercury mass with mercury signal (a) by direct injection and (b) following preconcentration with an Au/SiO_2 mercury cartridge



Mercury recovery of an Au/SiO_2 mercury cartridge in a SO_2-containing stream. The mercury recovery was calculated based on the linear regression of Y=36948þ1502X, where X is the mass of mercury (in picograms) and Y is the peak area (given in units of mVs), with a correlation coefficient of 0.9996. The synthetic flue gas contained 892 ppm SO2, 5.98% O2, 12.0% CO2, and the balance was N2.



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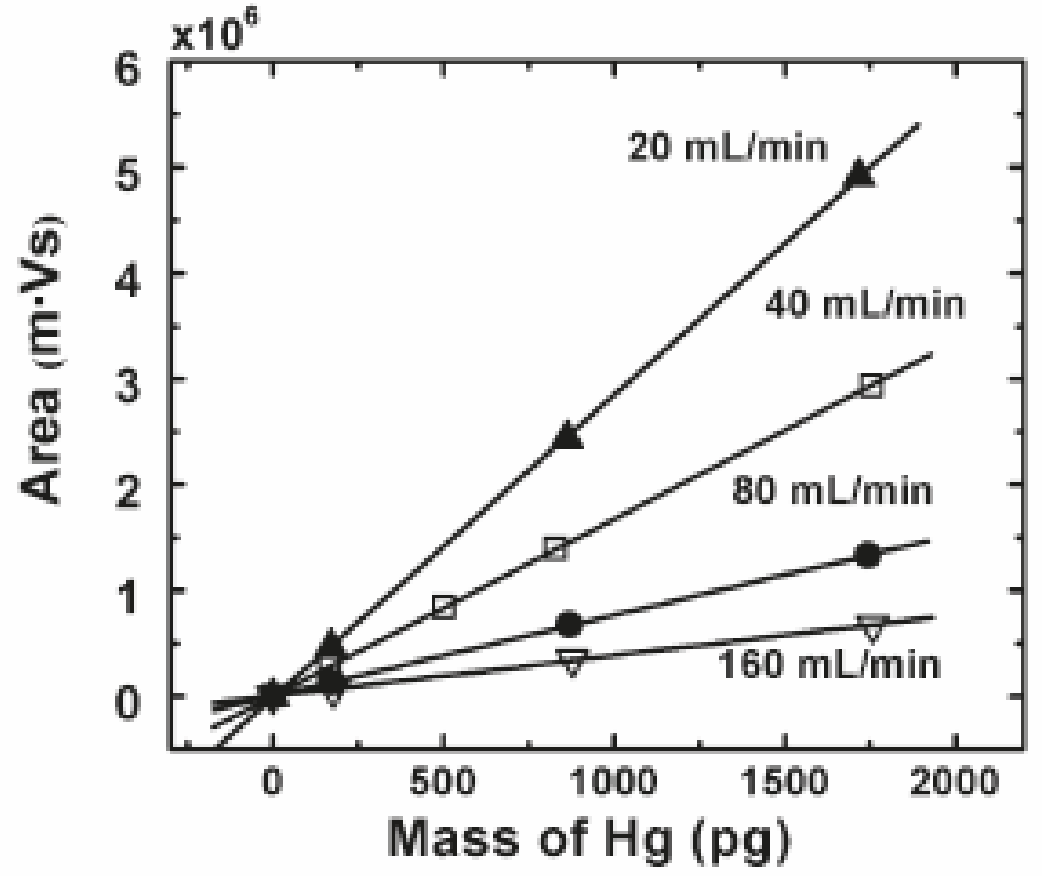
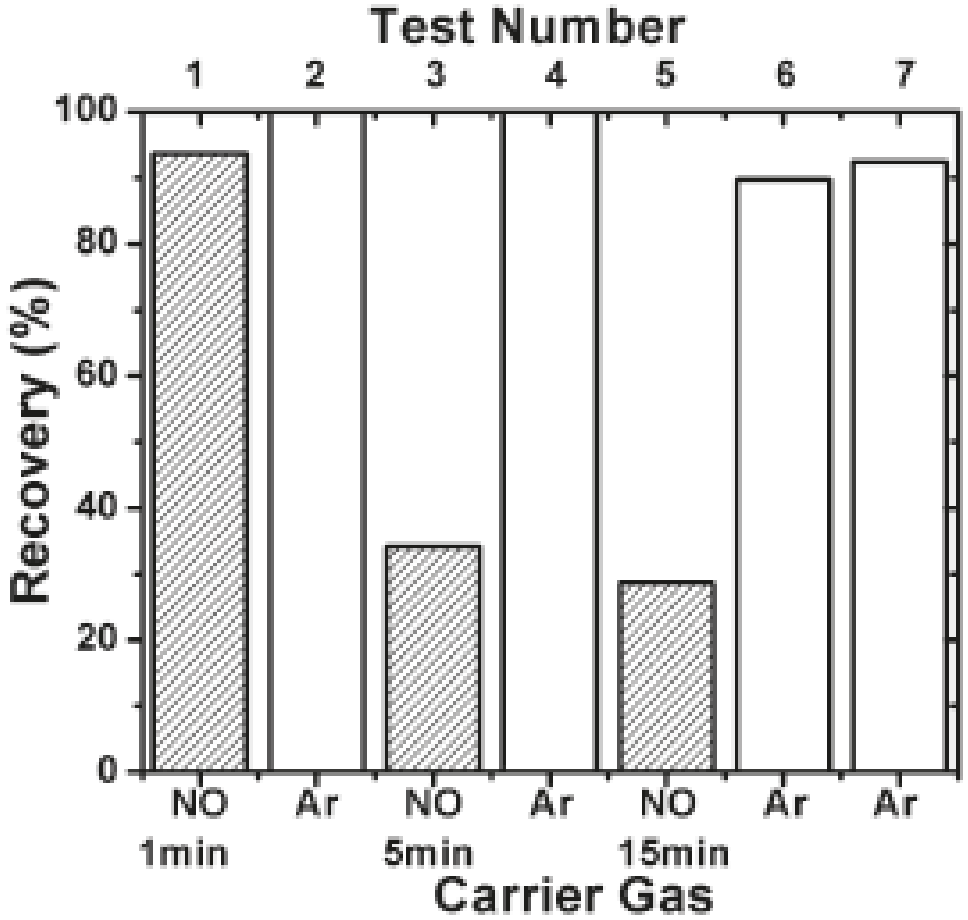
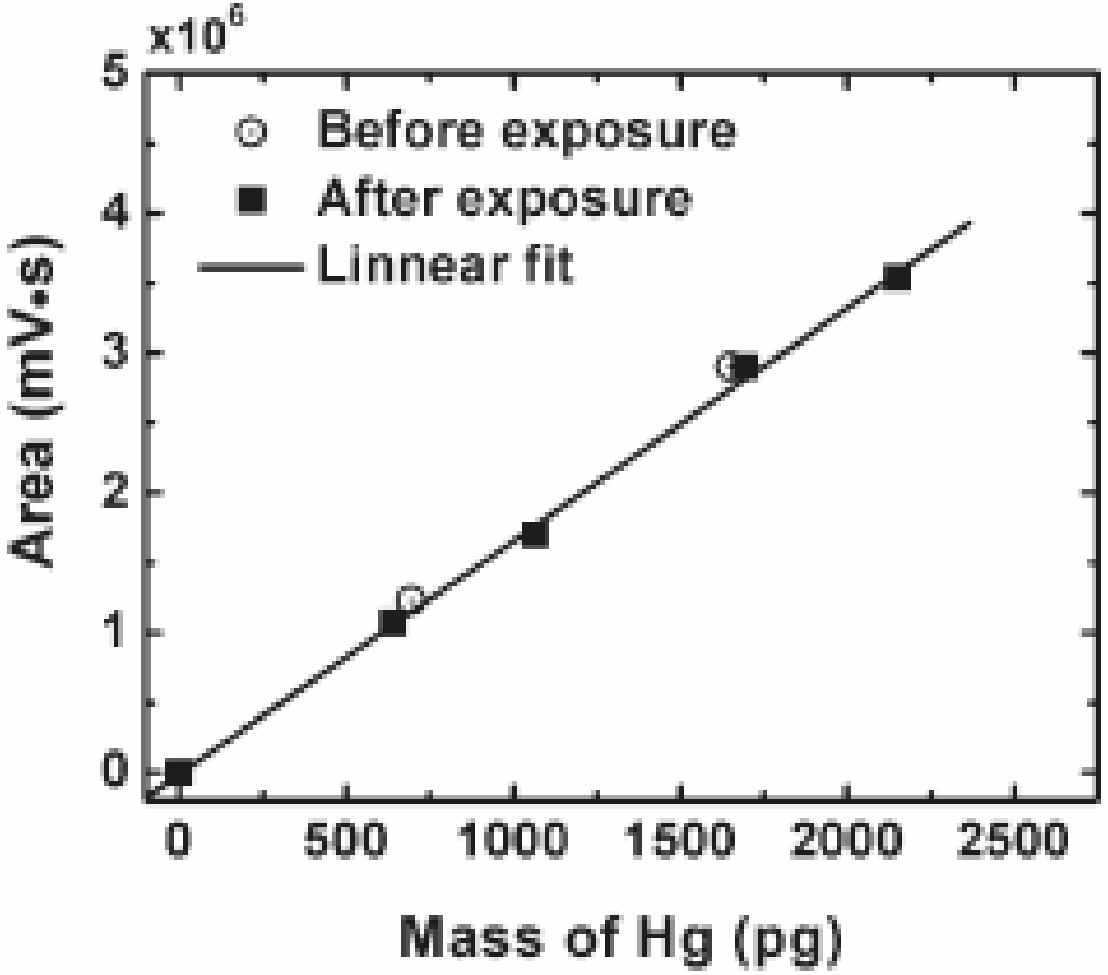


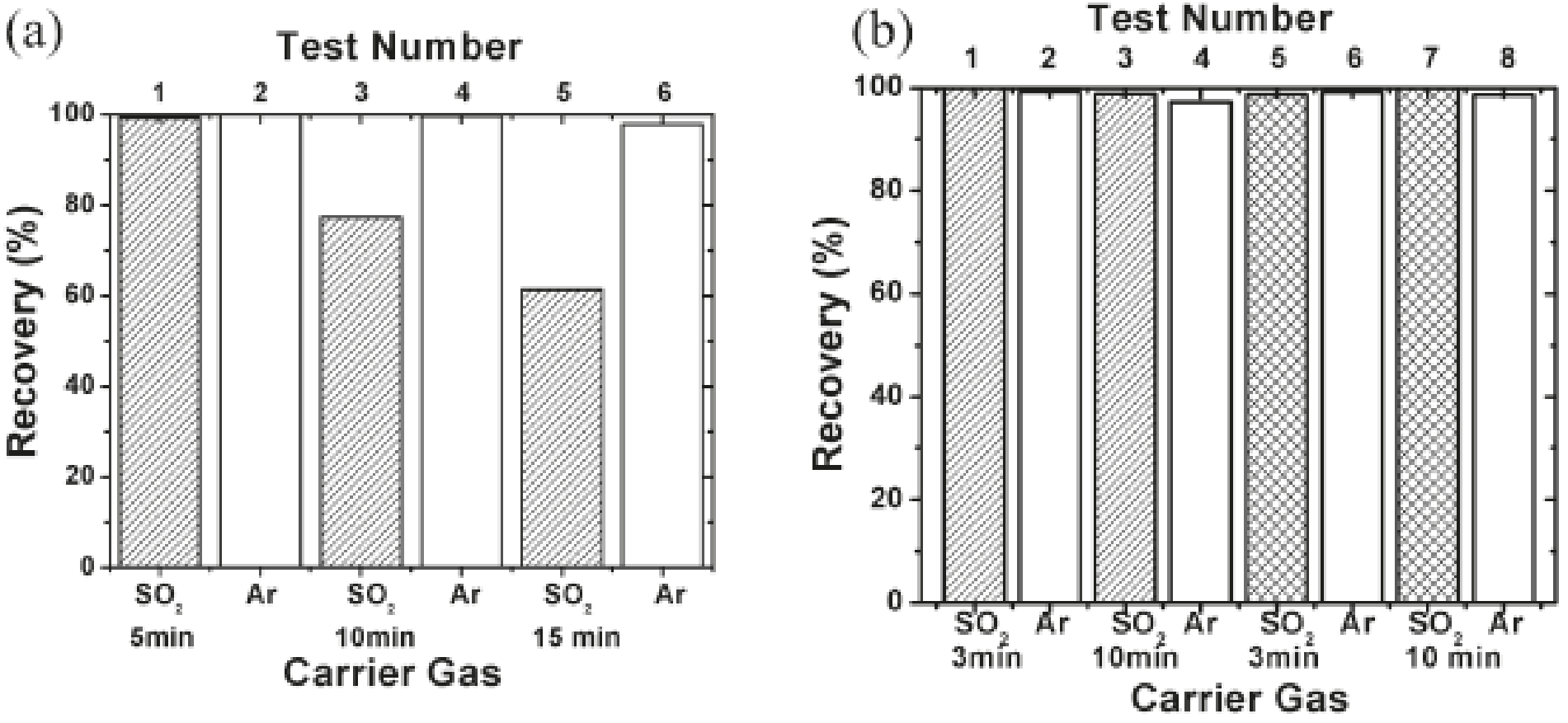
Correlation of mercury amount with peak area by an Ag/MC mercury cartridge at four different flow rates



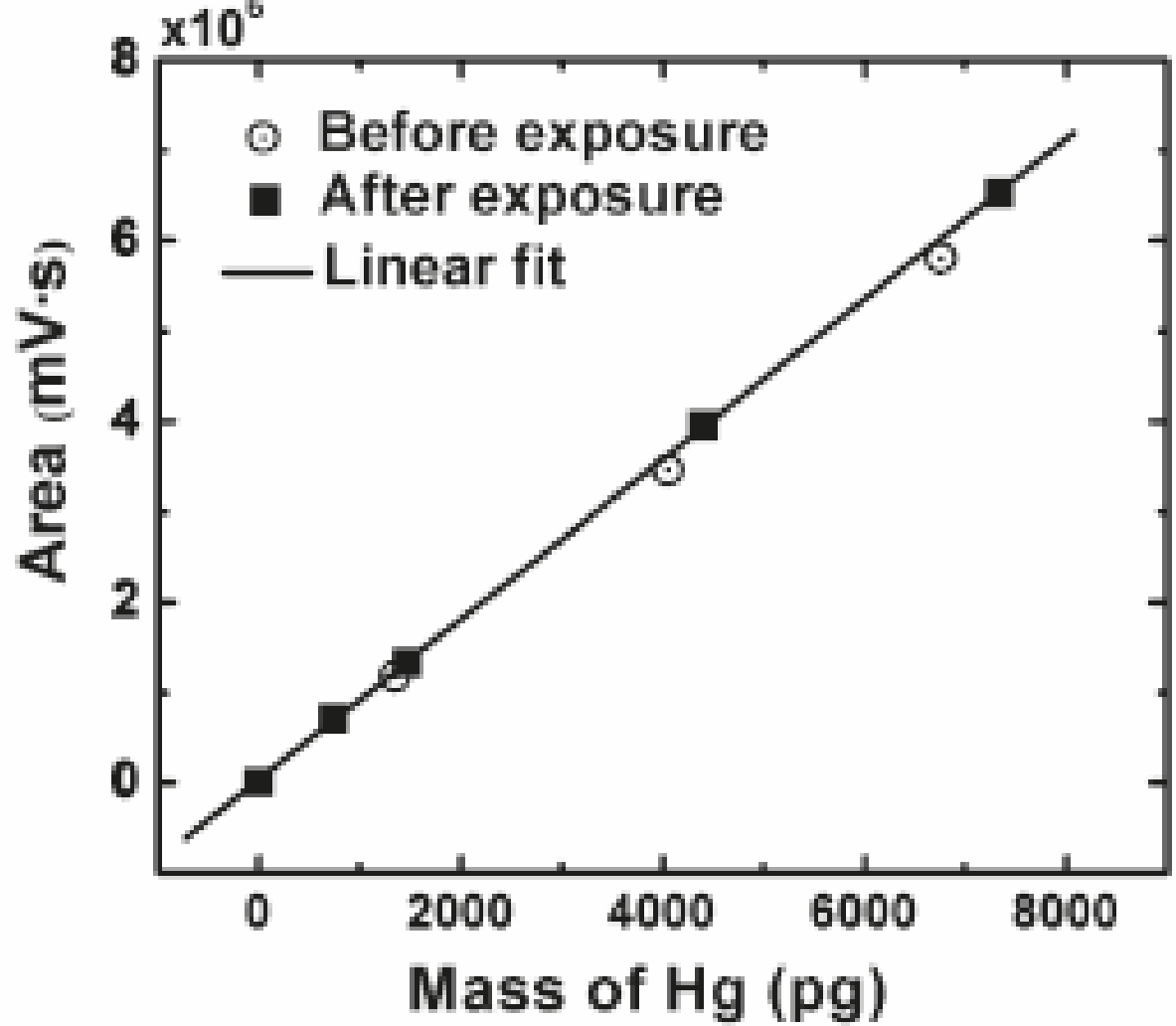
Mercury recovery of Ag/MC mercury cartridge in NOcontaining(318 ppm) N_2 stream with Ag/MC of 50 mg for test 1 to test 6 and 223 mg for test 7 to test 10. The mercury recovery was calculated based on the linear regression of Y=-26460 þ 1414X, where X is the mass of mercury injected in pg and Y is the peak area in mV3 s with a correlation coefficient of 0.9999



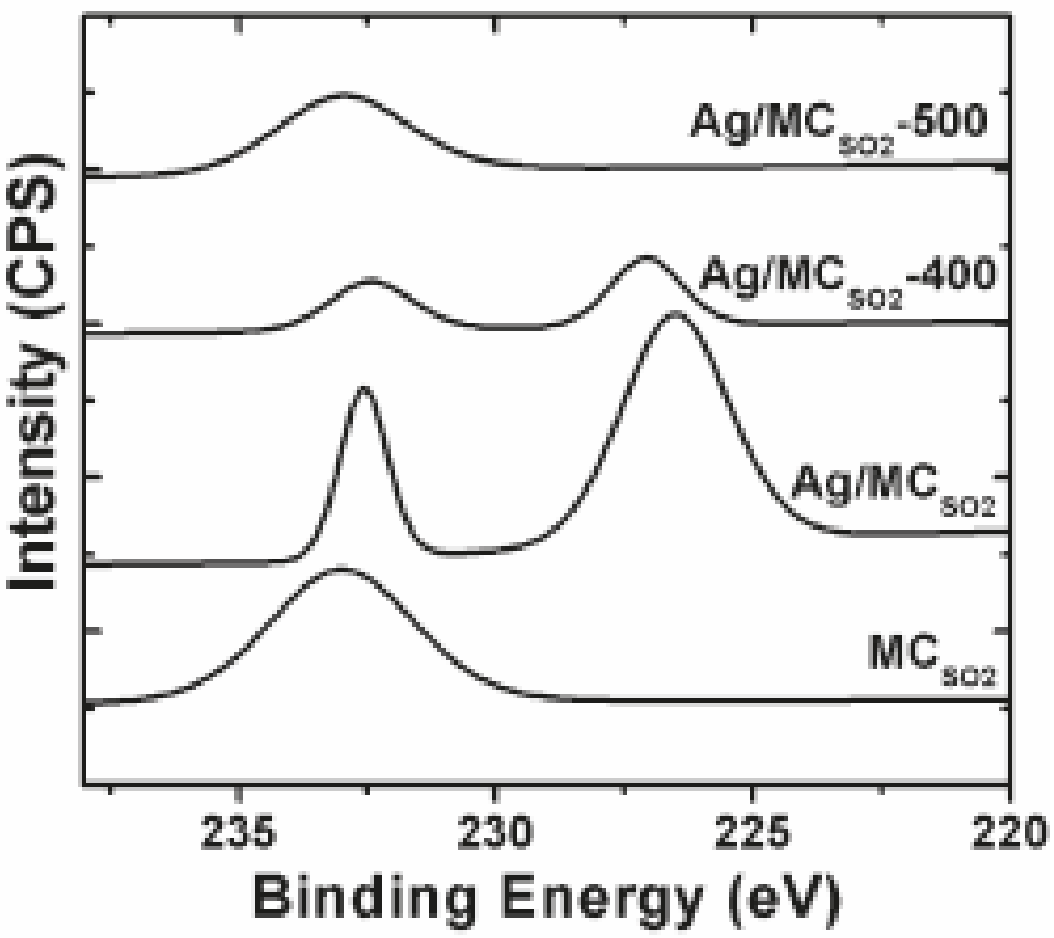
Effect of NO on mercury calibration using Ag/MC as mercury cartridge. The synthetic flue gas contained 318 ppm NO balanced by N_2. The mass of Ag/MC was 50 mg and the exposure time to NO was 5 min.



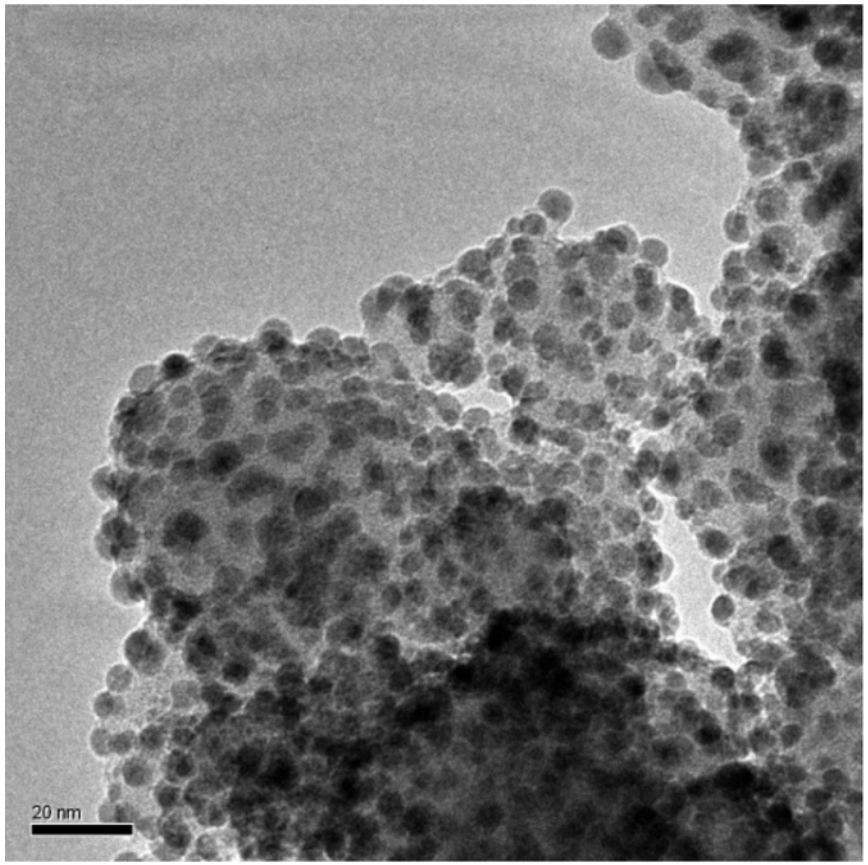
Mercury recovery in synthetic flue gas containing SO_2 of (a) Ag/MC mercury cartridge and (b) Ag/MC mercury cartridge coupled with an SO_2 scrubber. Hg recovery was calculated based on the linear regression of Y=7035þ2379X, where X is mass of mercury in pg and Y is peak area of mercury in mV.s with correlation coefficient of 0.9997.



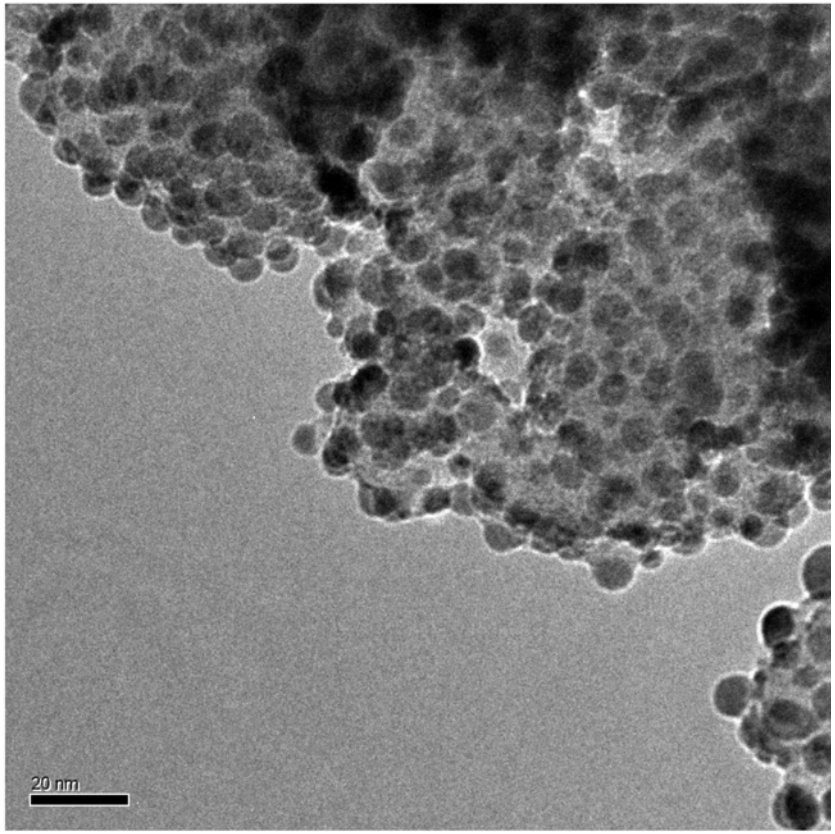
Calibration curve of mercury amount with mercury signal from Ag/MC in the presence of SO2 and NO. The exposure time to the synthetic gas was 5 min. The linear regression is Y=56883+800X, where X is the mass of mercury injected in pg and Y is the area of the peak in mV. s with linear correlation coefficient of 0.9995



XPS of S_{2s} peaks of MC and Ag/MC after SO_2 treatment (MC) at various temperatures



(a)



(b)

TEM images of Ag/MC (a) before and (b) after 5 h of 1500 ppm SO_2-containing stream exposure at 150 C.

Conclusions

- The mercury cartridge developed and fabricated in this study improves the linearity of response between mercury mass and mercury signal for a cold vapor atomic fluorescence absorption spectrophotometry (CVAFS) system.

- The presence of both NO and SO_2 affected Au/SiO_2 mercury cartridge performance. In contrast, the effect of NO was absent for an Ag/MC mercury cartridge, as long as the exposure time between the two consecutive measurements was controlled within a reasonable time frame.

- Using soda lime as a disposable SO_2 scrubber upstream of Ag/MC resulted in a novel mercury cartridge, which improved the accuracy of mercury measurement by CVAFS.

Thank you !