Simultaneous Removal of NO_x and Hg in a Low Temperature Selective Catalytic and Adsorptive Reactor

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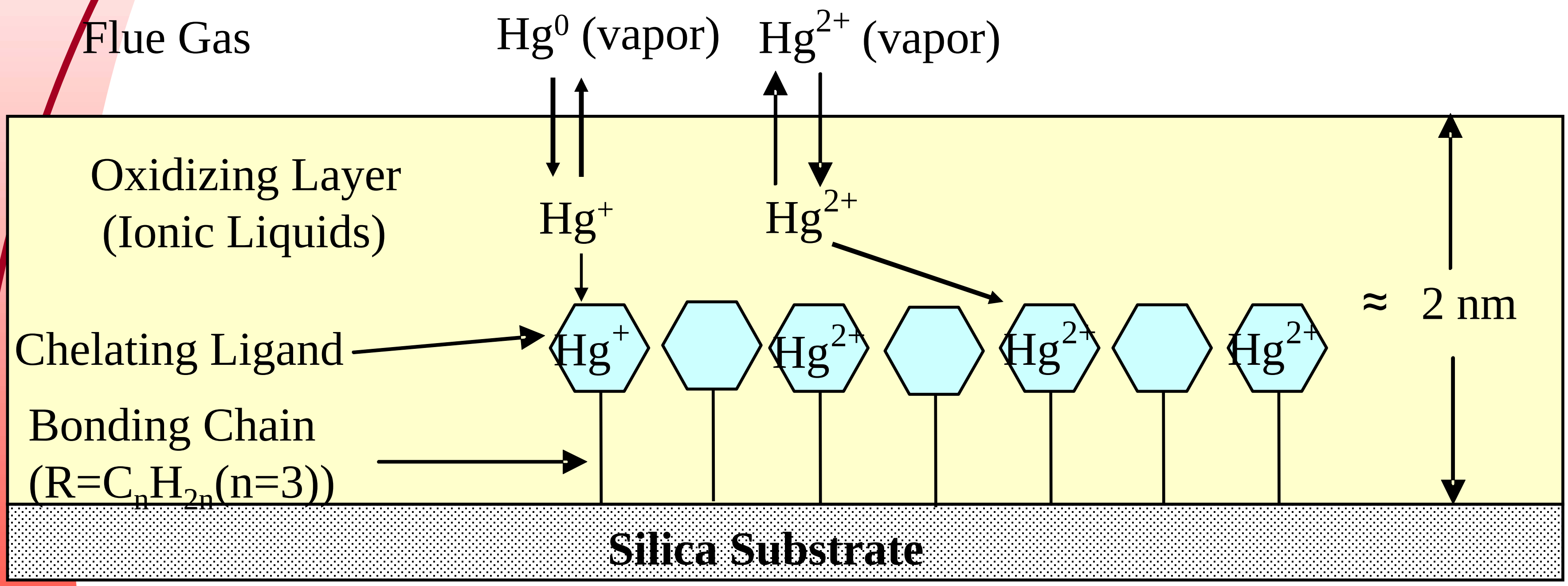
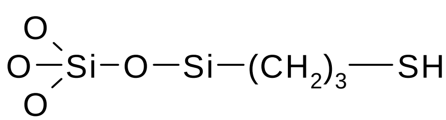
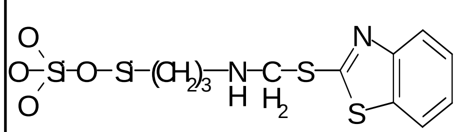
Thermally Stable Chelating Adsorbents for Hg^{2+} and Hg^0 Capture

Adsorption Group
Lei Ji, Stephen W. Thiel & Neville G. Pinto

Chelating Adsorbent



Chelating Ligands

Chelating Ligand	Chemical Structure	Active Sites Concentrartion	BET Area (Support+ Ligand)	Threotical Hg^{2+} Ligand Capacity	T_{max}
		mmol/g	m^2/g	(mg/g)	^oC
Pure Silica	-	-	283		-
MPTS (3-mercaptopropyltrimethoxysilane)		0.58	259	**117** (1:1)	200
APTS-MBT (Mercaptobenzothialzole)		0.22	245	**22** (2:1)	190

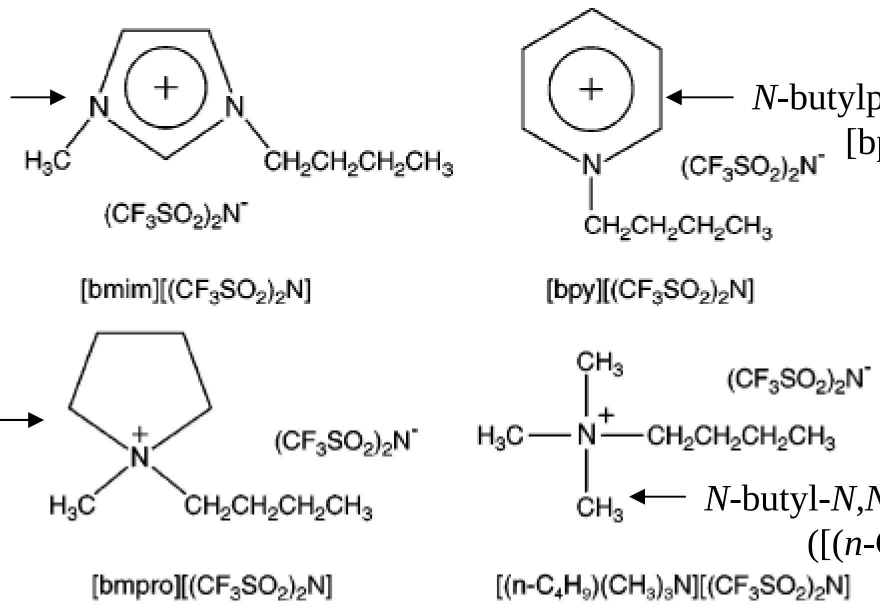
Ionic Liquids Screened

1-butyl-3-methylimidazolium
[bmim]

N-butylpyridinium
[bpy]

N-butyl-*N*-methylpyrrolidinium
[bmpro]

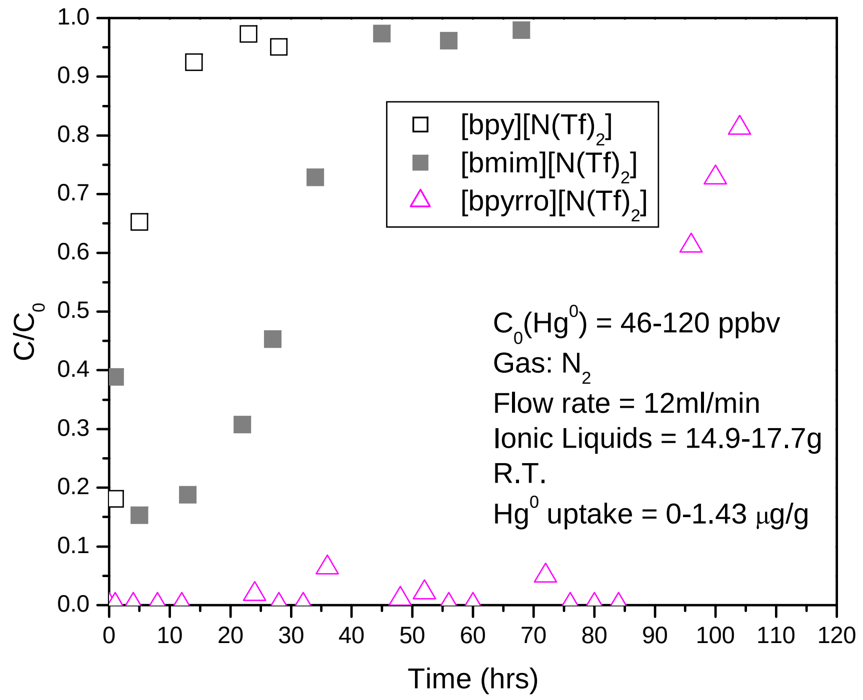
N-butyl-*N*,*N*,*N*,-trimethylammonium
([(*n*-C4H9)(CH3)3N])



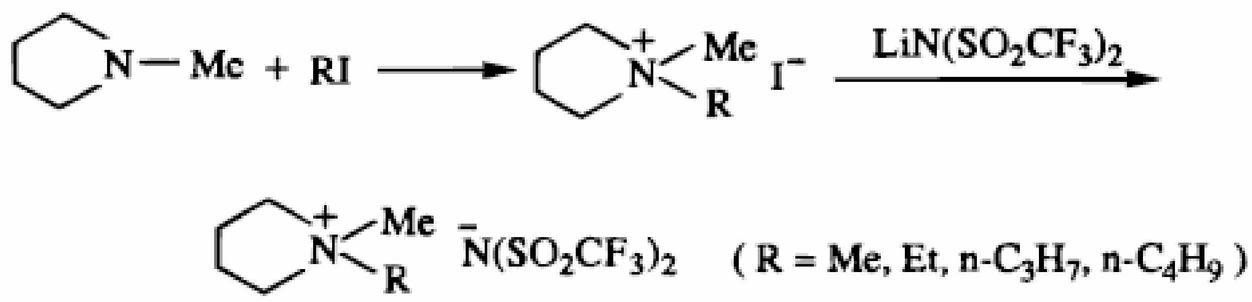
Same Anion: bis(trifluoromethane sulfonyl)imide
$[(CF_3SO_2)_2N]$

Tokuda et al., J. Phys. Chem. B 2006, 110, 6, 2833-2839

Effect of Cation of Ionic Liquids Coating on Mercury Capture

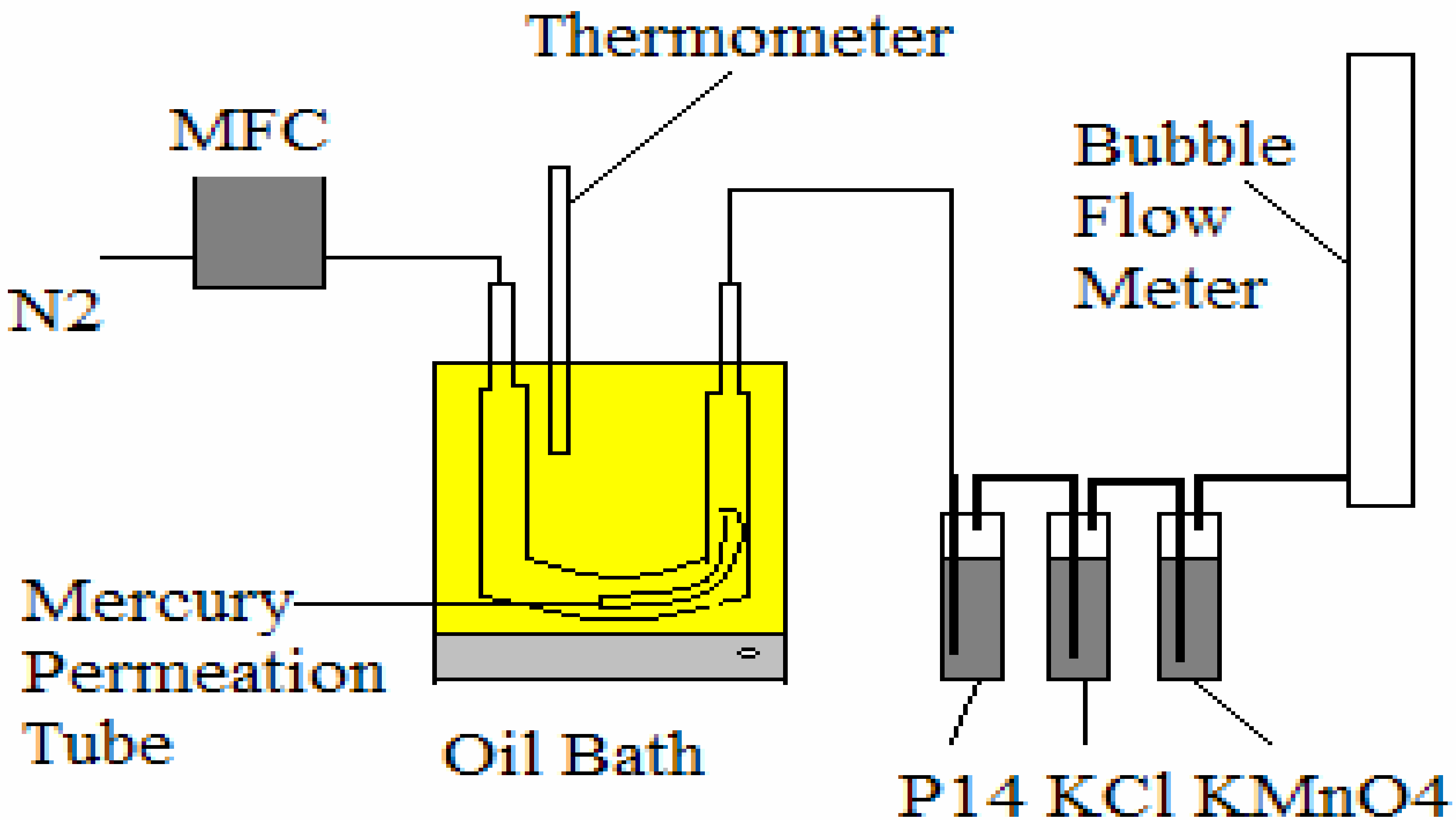


P_{14}: 1-butyl-1-methyl pyrrolidinium bis(trifluoromethane sulfonyl)imide

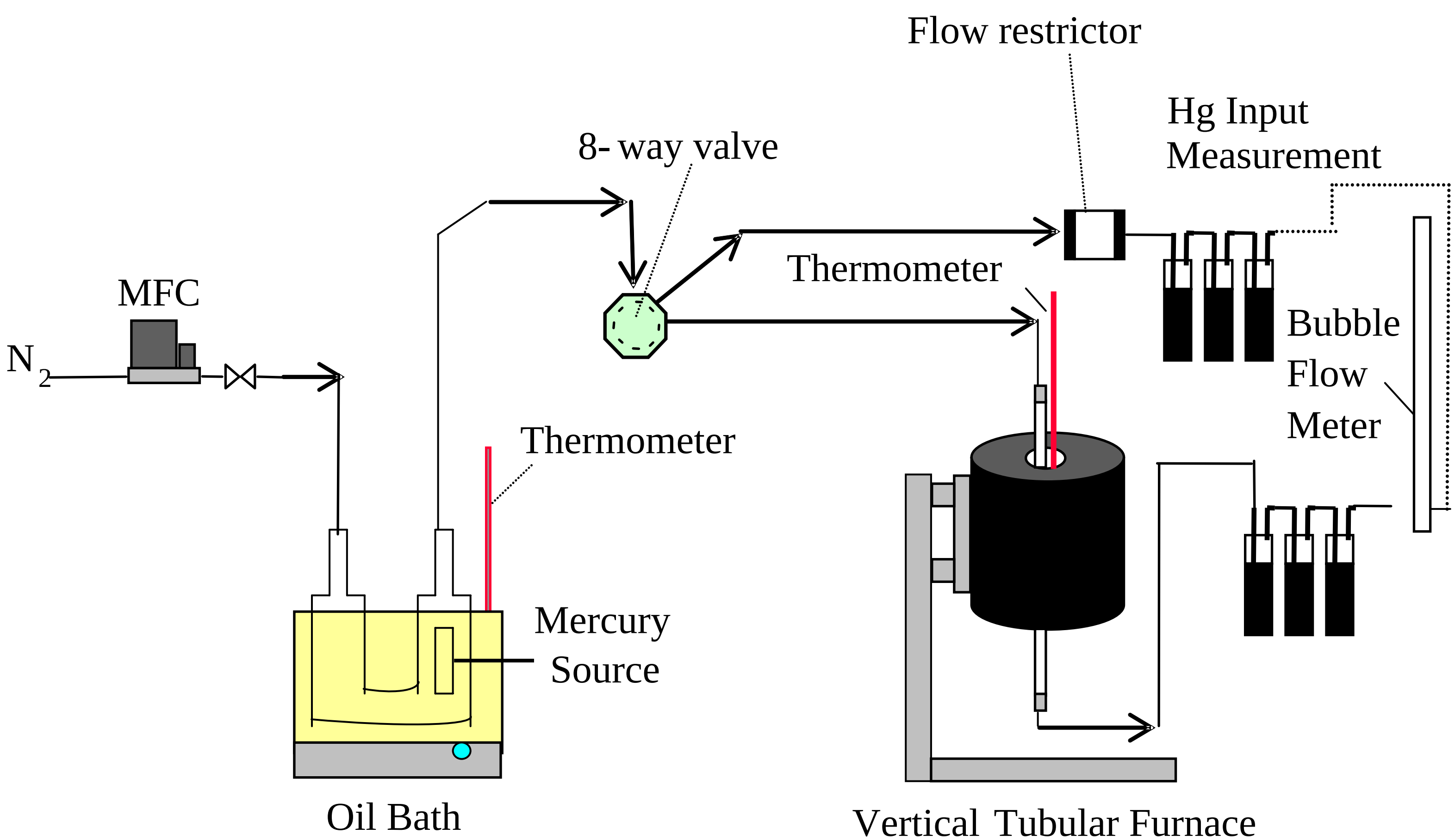


MacFarlane et al., J. Phys. Chem. B 1999, 103, 6, 4164-4170

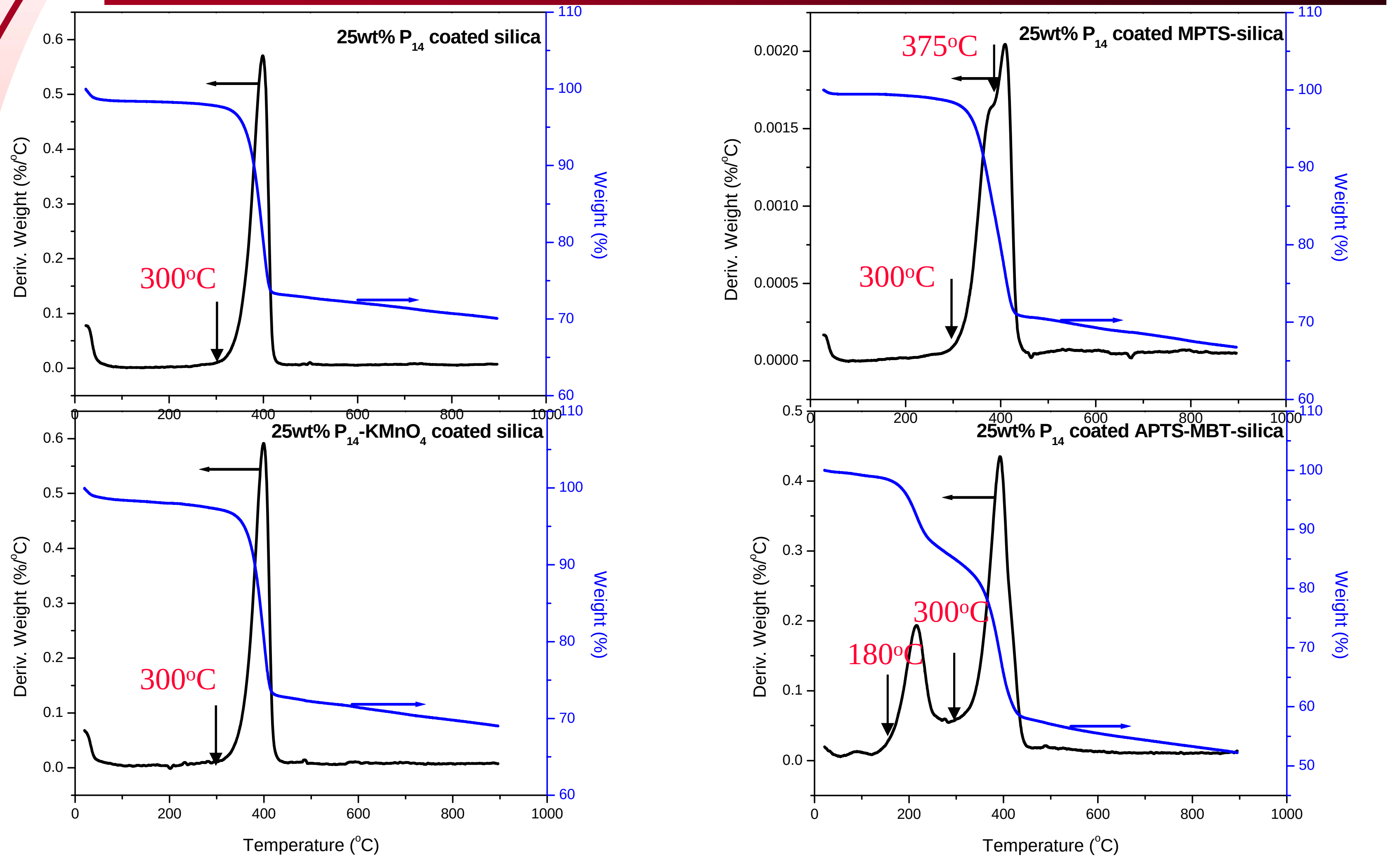
Apparatus (Bulk Phase)



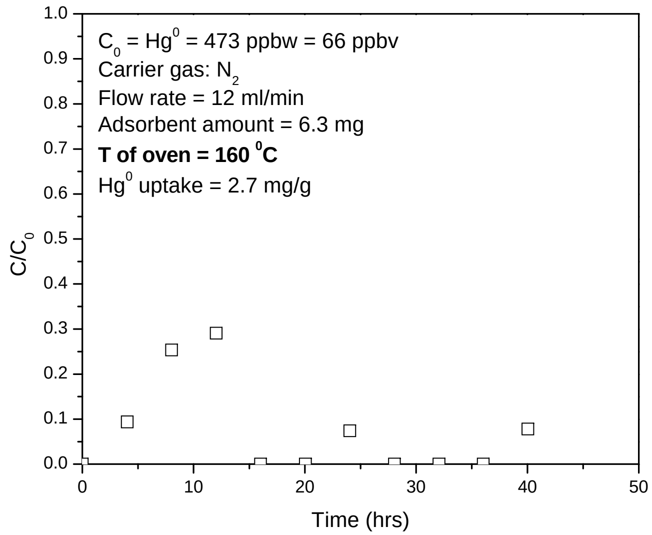
Apparatus (Fixed Bed)



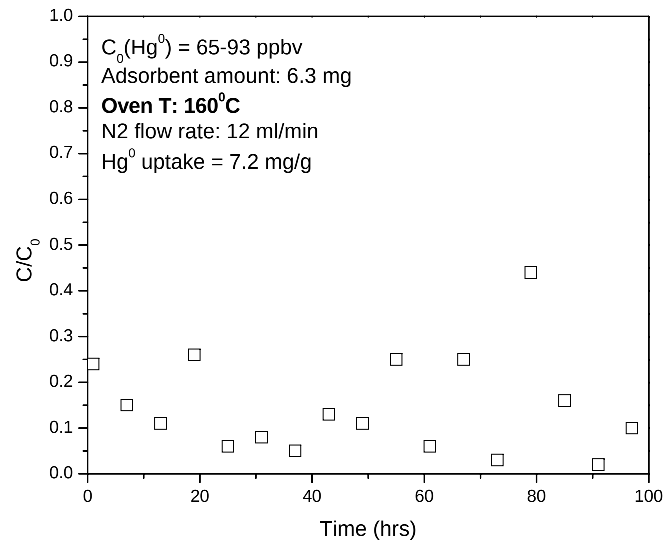
Thermal Stability



Mercury Capture Performance (Fixed-Bed at 160°C)



25 wt% P_{14} coated silica



25 wt% P_{14}-$KMnO_4$ coated MPTS-silica

Summary

- P_{14} and P_{14}-$KMnO_4$ are promising ionic liquids for Hg^0 capture
- P_{14} and P_{14}-$KMnO_4$ are stable up to 300^{o}C
- High Hg^0 uptake at 160^{o}C, several times higher than that of activated carbon
- Very rapid Hg^0 uptake, 0.04 seconds empty bed gas residence time
- The MPTS (3-mercaptopropyltrimethoxy-silane) chelating ligand was found to be the most effective than APTS-MBT (mercaptobenzo-thialzole)

TiO_2 Supported MnO_2 DeNOx Catalyst for Hg^0 Capture

Adsorption Group
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Catalysis Group
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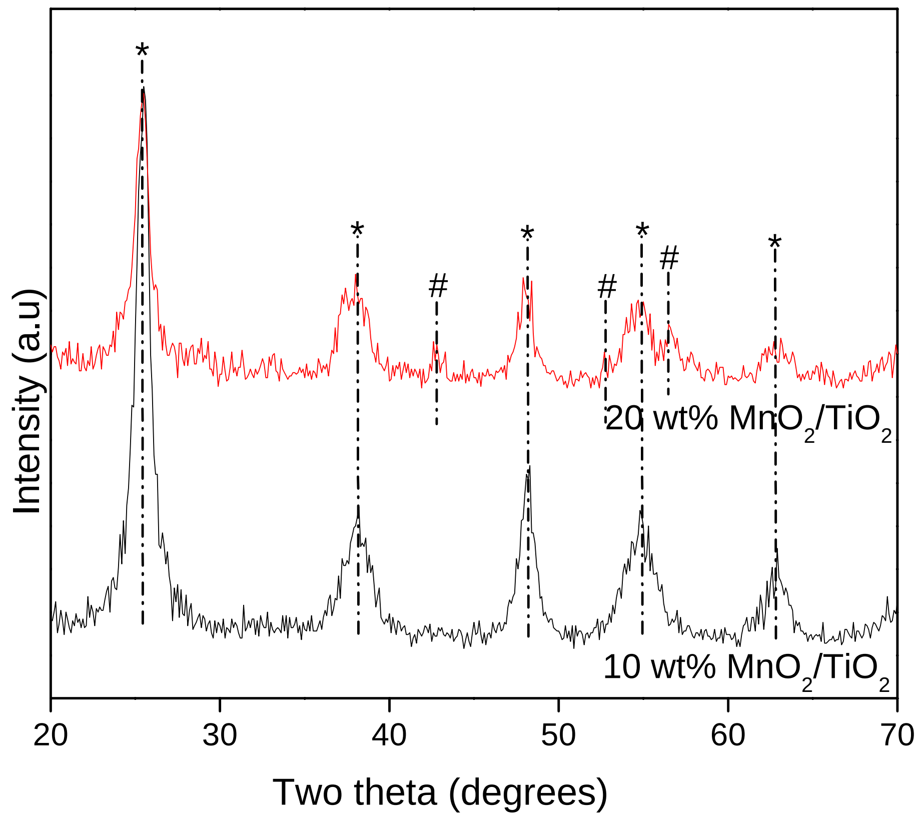
BET Surface Area and Total Acidity of MnO_2/TiO_2 Catalysts

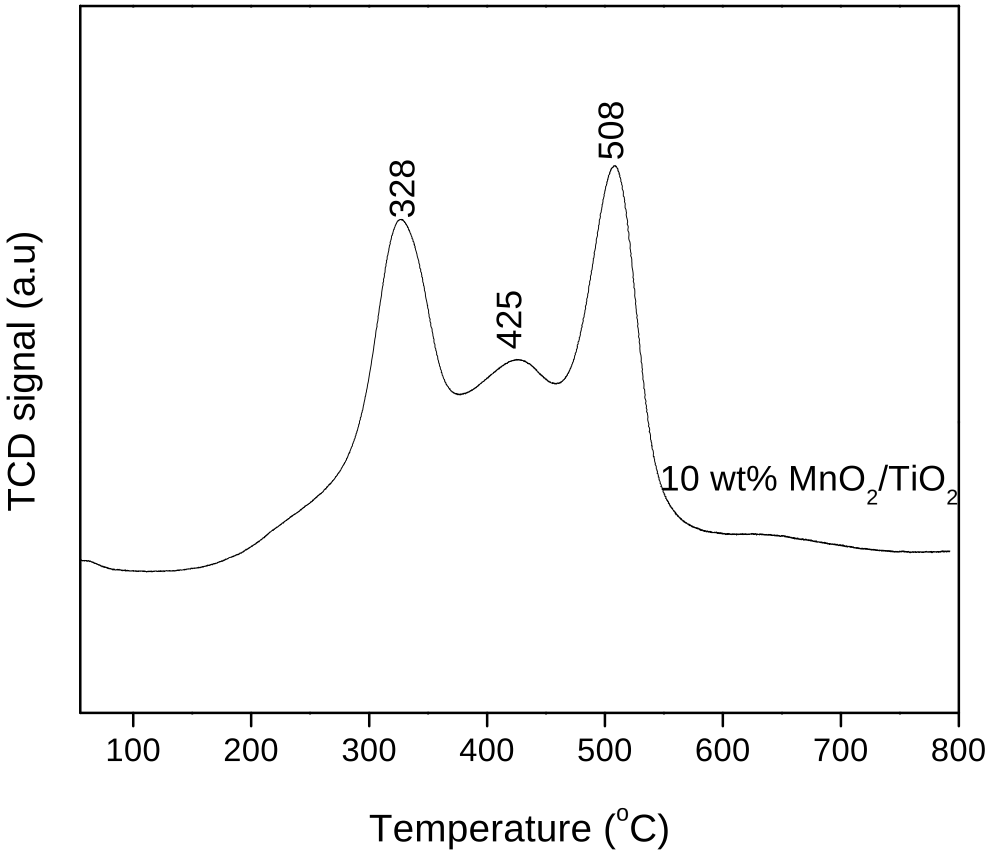
Sample	**BET surface area** (m^2/g)	**Total Acidity** (ml g^{-1})
TiO_2	309	-
10 wt% MnO_2/TiO_2	236	21
20 wt% MnO_2/TiO_2	198	23.2

Powder XRD patterns for MnO_2/TiO_2 SCR catalysts



- ❖ (*) anatase TiO_2
- ❖ (#) MnO_2

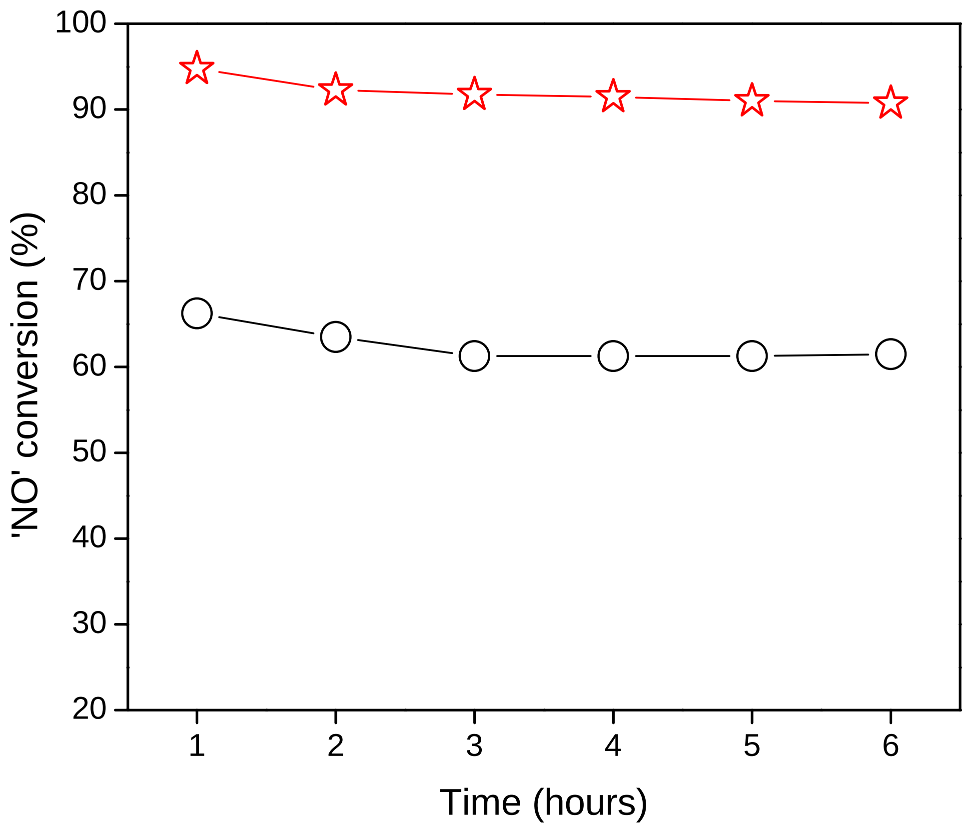
TPR pattern of 10 wt% MnO_2/TiO_2 catalyst



$MnO_2 \rightarrow Mn_2O_3 \rightarrow$

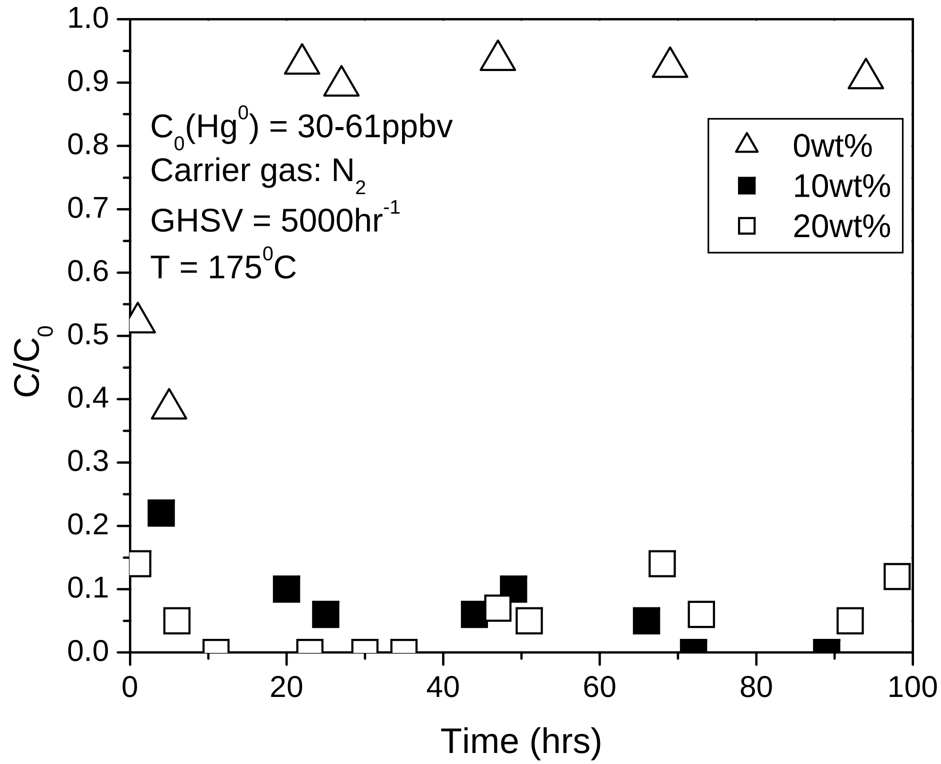
$Mn_3O_4 \rightarrow MnO$

SCR of NO using CO as a reductant over 10 wt% MnO_2/TiO_2 catalyst



(○) 175 ^{0}C; (☆) 200 ^{0}C;
NO = CO= 400 ppm;
Oxygen = 2 vol%

Influence of MnO_2 Loading on Mercury Capture Behavior



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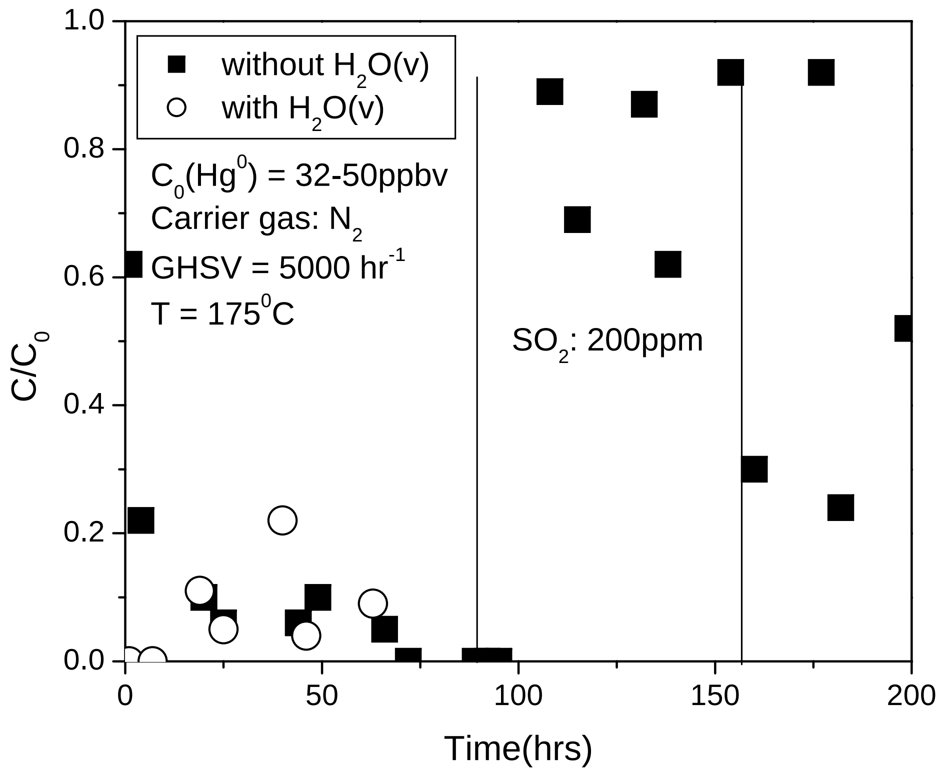
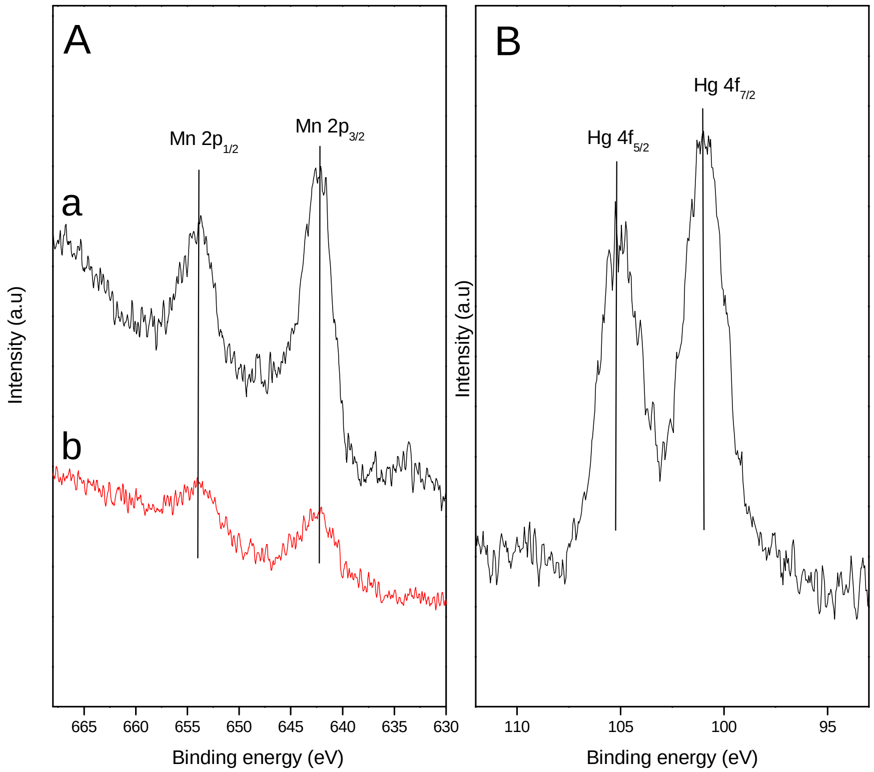
Effect of SO_2 and Water Vapor on Mercury Capture Behavior using 10wt% MnO_2 Catalyst

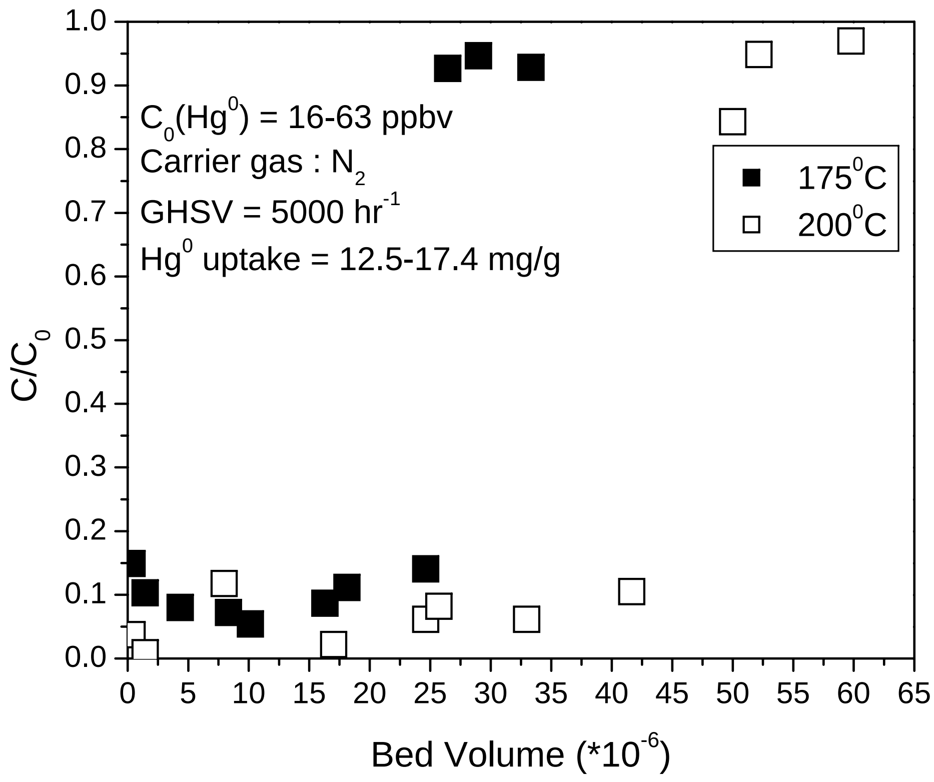


XPS spectra of fresh and spent samples of 10 wt% MnO_2/TiO_2 catalyst

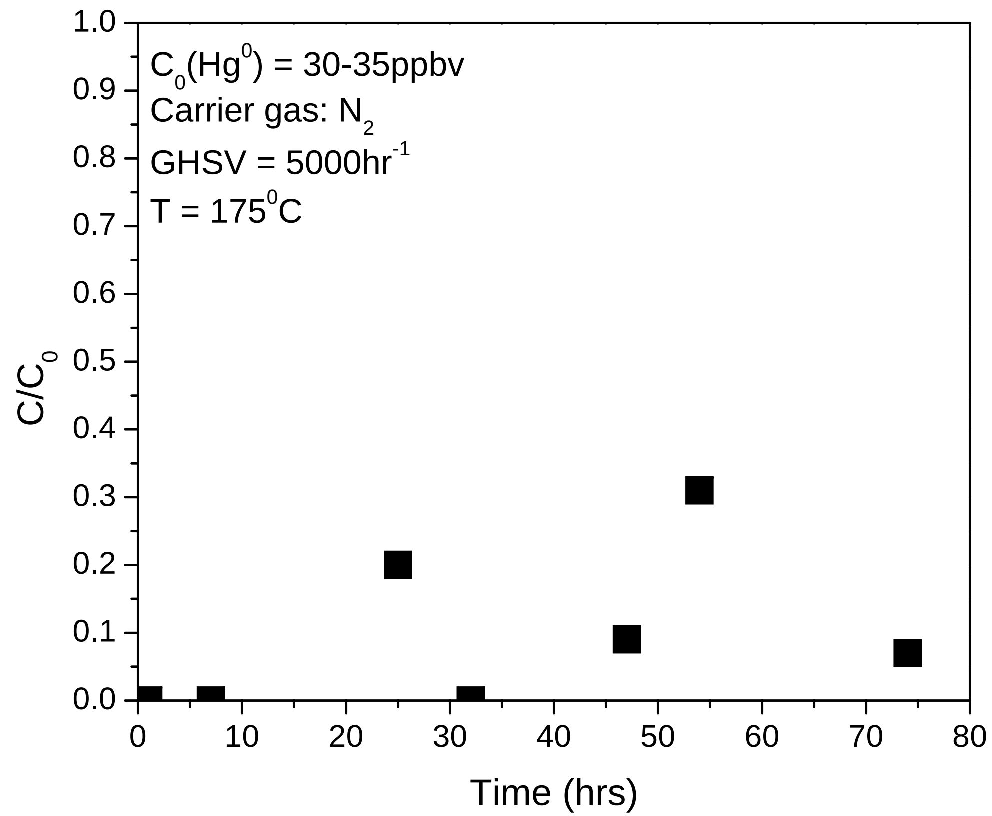


(A) Mn 2p XPS spectra for (a) fresh and (b) spent catalysts
(B) Hg 4f XPS spectrum for spent catalyst

Effect of Bed Temperature on Mercury Capture Behavior using 10wt% MnO_2 Catalyst



Mercury Capture Behavior using 20wt% MnO_2 Catalyst after NOx Conversion



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Summary

- Remarkable mercury capacity was observed at 200 ^{0}C and inert atmosphere
- MnO_2/TiO_2 catalysts are water tolerant. SO_2 has a negative effect on mercury capture
- Manganese loading and bed temperature were found to be important factor for mercury capture
- XPS results indicate that MnO_2/TiO_2 are covered with mercury

Acknowledgements

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- Dr. James Boerio for assistance with the XPS analysis.
- University of Cincinnati