Chemical Engineering Journal 200–202 (2012) 91–96

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

The adsorption mechanism of elemental mercury on CuO (1 1 0) surface

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HIGHLIGHTS

- ▶ Density functional theory is used to investigate the adsorption mechanism.
- ▶ The surface is represented by periodic model, and different sites are considered.
- ▶ Adsorption energies, bond length, and bond populations are calculated.
- ▶ The electronic structural changes upon adsorption are studied.

ARTICLE INFO

Article history:
Received 20 March 2012
Received in revised form 7 June 2012
Accepted 7 June 2012
Available online 16 June 2012

Keywords:
Mercury
Coal combustion
Adsorption
CuO

ABSTRACT

Understanding the impact of CuO in selective catalytic reduction (SCR) process for elemental mercury removal will broaden the applicability of SCR system in Hg removal strategies. First principles quantum mechanical methods based on density functional theory were used to investigate the adsorption mechanism of Hg on CuO (1 1 0) surface. The CuO (1 1 0) surface was represented by a periodic model, and different adsorption sites were considered. The electronic structural changes upon adsorption were also studied to better understand the surface reactivity. The results show that elemental mercury binds weakly to the O-terminated CuO (1 1 0) surface, which indicates a physisorption mechanism. On the contrary, Hg is strongly adsorbed on the Cu-terminated CuO (1 1 0) surface and chemisorption is the likely adsorption mechanism. The adsorption of Hg on CuO (1 1 0) surface is mainly by the Cu-terminated mode. Cu_{sub} top is the most advantageous adsorption site with an adsorption energy of −116.76 kJ/mol. In addition, bond population analysis indicates that Hg atom preferably adsorbs on CuO (1 1 0) surface with the bonding of Cu atoms. According to the calculation of the partial density of states of the surface atoms, strong mercury interactions with the surface cause a significant overlap between the d-state of mercury and the s-states of Cu.

1. Introduction

Mercury is a major atmospheric pollutant because of its toxicity, ability to be transported in the environment, adverse effects on the ecosystem and negative effects on human health and aquatic life [1]. According to the recent report by the United Nations Environment Programme (UNEP), burning of fossil fuels (primarily coal) is the largest single source of emissions from human sources, accounting for about 45% of the total anthropogenic emissions [2]. Hence, the reduction of mercury emissions in coal-fired power plants has become a matter of great public concern.

The forms of mercury are of particular importance for removing mercury [3]. Mercury in the flue gas occurs primarily in three forms: oxidized gaseous compounds (Hg^{2+}), gaseous elemental (Hg^0) and particle-bound (Hg_p). Conventional air pollution control devices such as electrostatic precipitator (ESP) or fabric filter (FFs) can collect Hg_p efficiently together with fly ash particle [4]. Hg^{2+} is soluble in water and has a tendency to stick to particulate matter. This physicochemical property of Hg^{2+} makes it easily to be removed by ESP, FFs, or flue gas desulfurization (FGD) [5]. On the contrary, Hg^0 is more difficult to remove than Hg^{2+} because Hg^0 is less reactive, highly volatile and water insoluble [6]. Thus a low cost Hg^0 adsorption/oxidation process that can be applied for flue gas treatment is needed.

Considerable studies have been conducted to find an effective approach for removing Hg^0 from coal-fired flue gas. Researchers have demonstrated that selective catalytic reduction (SCR) catalysts for NO_x reduction are available for converting Hg^0 to Hg^{2+} which is easily to be removed [7–9]. It offers a cost effective option for mercury control in coal-fired power plants. EPA regarded it as "co-benefit" controls for mercury emissions achieved by reducing

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http://dx.doi.org/10.1016/j.cej.2012.06.025