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Adsorption and catalytic oxidation of elemental mercury over regenerable magnetic Fe—Ce mixed oxides modified by non-thermal plasma treatment

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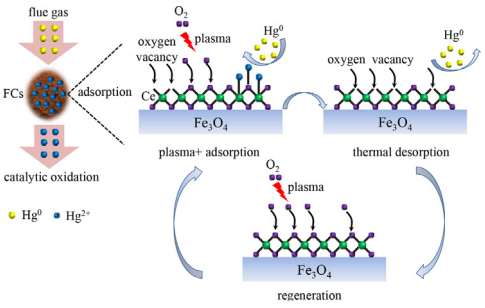
HIGHLIGHTS

- Non-thermal plasma is proposed to modify magnetic Fe—Ce oxides for efficient Hg^0 removal.
- The Hg^0 removal performance is substantially increased after non-thermal plasma treatment.
- The Hg^0 removal mechanism was revealed by XPS, Hg-TPD and pseudo-second-order model.
- The spent Fe—Ce oxides can be effectively regenerated via non-thermal plasma treatment.

GRAPHICAL ABSTRACT



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ABSTRACT

This study proposes the novel application of non-thermal plasma treatment to improve the oxidation capacity of regenerable magnetic Fe—Ce mixed oxides (FCs) for the efficient removal of elemental mercury (Hg^0) from coal combustion flue gas. Sample characterization shows that the textural property, crystalline phases, and magnetic property of FCs undergo no obvious changes after plasma treatment. But greater Ce^{4+} concentration and richer lattice oxygen are generated on the treated FCs. The treated FCs exhibit far better Hg^0 removal performance compared to raw FC. The effects of treatment time (0–20 min), reaction temperature (100–250 °C), and flue gas components (SO_2, NO, O_2, HCl and H_2O) on Hg^0 removal performance are also discussed. Both Hg^0 adsorption capacity and adsorption rate evaluated at 150 °C for the treated FCs are extremely close to those obtained with a commercial activated carbon manufactured specifically for mercury removal from flue gas. Furthermore, the Hg^0 removal mechanism is proposed for the treated FCs. The treated FCs include separate active sites for Hg^0 adsorption and catalytic oxidation. Ce^{4+} species with greater oxidation state and lattice oxygen are largely consumed during the Hg^0 removal process. However, these components are replenished by subsequent non-thermal plasma treatment. Finally, the spent FCs can be effectively recycled through magnetic separation, thermal desorption, and non-thermal plasma treatment.

1. Introduction

Mercury has become a severe threat to human health worldwide because of its bioaccumulation, toxicity, and persistence in the environment [1,2]. Combustion flue gas emissions from coal-fired power plants are currently considered to be the main anthropogenic sources of mercury [3,4]. Three species of mercury are primarily found in coal combustion flue gas: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}), and particulate-bound mercury (Hg^P) [5–8]. Hg^{2+} and Hg^P can be effectively removed by currently existing air pollution control

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