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Experimental study on catalytic cracking of model tar compounds in a dual layer granular bed filter

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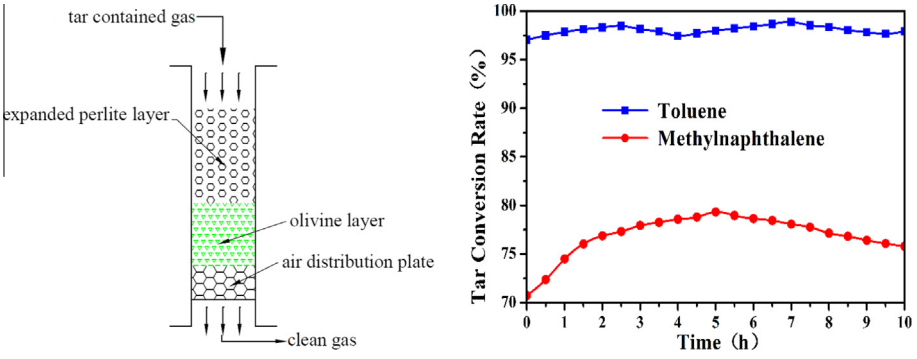
HIGHLIGHTS

- A novel technique for the simultaneous removal of particulates and tar is proposed.
- A tar removal catalytic filter based on a granular bed filter is investigated.
- Catalytic filter reaches 98.89% toluene and 79.33% methylnaphthalene conversions.

GRAPHICAL ABSTRACT



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ABSTRACT

To simplify the purification process of hot effluent gases produced by biomass and/or coal gasification, an integrated technique employing dual layer granular beds for the simultaneous removal of particulates and tar at high temperature (750–850 °C) has been proposed. Compared to the well studied process of particulate removal, investigations for the catalytic cracking of tar are inadequate. The present work conducted an experimental study of the catalytic cracking of tar based on two model compounds: toluene and methylnaphthalene. The experiments were conducted on a granular bed filter with an inner diameter of 40 mm and a height of 1000 mm. The catalytic cracking performances on an expanded perlite granule layer, an olivine granule layer, and a dual layer granular bed comprised of an upper layer of expanded perlite granules and a lower layer of olivine granules were investigated individually. It was found that the expanded perlite granule layer alone was effective for toluene cracking, and the efficiency was increased with increasing reaction temperature and gas residence time. The olivine granule layer provided excellent catalytic activity for toluene cracking. Furthermore, olivine granules exhibited significantly improved catalytic activity when calcinated at temperatures greater than 900 °C, or impregnated with NiO. The conversion efficiencies of toluene and methylnaphthalene attained values as great as 98.89% and 79.33%, respectively, at a temperature of 800 °C over a dual layer granular bed comprised of a 150 mm thick upper layer of expanded perlite granules and a 50 mm thick lower layer of 3 wt% NiO impregnated olivine granules.

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1. Introduction

Due to the growing limitations in fossil energy resources and increasing global environmental concerns, the substitution of fossil energy sources by renewable sources is playing an ever-increasing role in the energy field. The high-efficiency and clean utilization of coal and the large-scale application of renewable biomass are considered to be two effective routes to meet the demand. Gasification is one of the main methods employed for clean and high-efficiency utilization of biomass and coal. However, this process inevitably produces solid particulates (ash, alkali metal compounds, char, etc.) and tar [1]. The presence of these by-products can affect the normal operation of downstream equipment, reduce gasification efficiency (the tar itself contains about 3% of the total biomass energy [2]), and result in the release of environmental pollutants [3–7]. Bridgwater et al. [8] and Ma et al. [9] reported that particulates and tar produced in high-temperature gasification processes have concentrations of 5–30 g/Nm^3 and 15–20 g/Nm^3, respectively. For most downstream devices, especially for turbines, the concentrations of tar and particulate in the inlet gases must be less than 50 mg/Nm^3 and 2.4 mg/Nm^3, respectively [10,11]. As a result, the removal of tar and particulates from hot effluent gases obtained in gasification processes is a fundamental concern, and is therefore a subject of great interest. The aim of the present study is focused on developing a high-efficiency, energy-saving simultaneous particulate and tar removal method for biomass or coal gasification processes.

Cleanup technologies for the syngas produced from coal/biomass gasification can be characterized according to two methods: cold gas cleanup and hot gas cleanup [12]. Cold gas cleanup technologies employ wet scrubbing by a liquid absorbent such as water, vegetable oil, diesel fuel, or engine oil [13–15]. Wet scrubbing techniques are simple, commonly used methods for tar and particulate removal [16]. However, the fact that tar tends to condense on the surface of ash particles during wet scrubbing is not favorable for tar processing. Additionally, the process of water scrubbing generates very large quantities of waste-water, resulting in secondary pollution. Similarly, scrubbing conducted using an oil-based absorbent will also generate secondary pollution, and further increase the cost and complexity of the process [13]. Heat waste is another obvious drawback of scrubbing processes. In contrast, hot gas cleanup has recently attracted increasing attention for the removal of tar and particulates because the method can reduce waste streams and increase the energy efficiency [17,18]. Tar can be removed efficiently either by strictly thermal or catalytic cracking methods at high temperature. The typical reaction temperature for strictly thermal processes is greater than 1000 °C [19,20], and this technology has been abandoned owing to the increasing cost of more sophisticated gasification equipment, the very large consumption of heating energy, and the considerable production of soot [20–22]. In contrast, catalytic cracking for tar removal operates under low temperatures compared to that of a strictly thermal process. Tar conversion efficiency for catalytic cracking is high at temperatures ranging from 750 to 850 °C with the use of a suitable catalyst such as dolomite, olivine, iron, or nickel-based catalysts [11,20,23]. Catalytic reforming is eminently suitable for tar removal because the technology can convert tar into environmentally friendly and more readily removable forms, or even into useful gases [24,25], which eliminates the production of organic pollutants and the need for tar recycling. Two methods for catalytic removal of tar have been reported: primary removal, for which the catalyst is located within the gasifier; secondary removal, where the catalyst is located outside of the gasifier [26]. Clearly, the secondary removal method is more efficient, economical, and easier to control [27]. To prevent fly ash and dust from clogging the catalyst channel in the tar reformer, high temperature filtration of hot gas is necessary. However, the upper temperature limit for which existing preliminary particulate removal equipment function reliably ranges from 350 °C to 500 °C [28]. Therefore, due to this limitation, the high temperature gas from a gasifier must be cooled to within this proper functioning temperature range for conducting preliminary particulate filtration in a typical gas purification process, as shown in Fig. 1A. This is similar to the Clean Hydrogen-rich Synthesis Gas (CHRISGAS) project [29]. The gas is then heated to above 700 °C before entering the catalytic tar cracker for tar removal. The gas temperature is again lowered to 150–200 °C before it enters the Venturi wet scrubbers for secondary particulate filtration to remove residual particulates and any other harmful substances. Finally, after water washing purification, the gas can be applied in a variety of terminal equipment. This process involving repeated cooling and heating is obviously complex and consumes an excess amount of energy because the waste thermal energy cannot be fully used. Therefore, high temperature filtration and tar removal are the most challenging processes in gasification [25].

The most effective and economical method for the purification of the effluent of gasification processes is the simultaneous removal of particulates and tars at high temperature by combining hot gas filtration and catalytic tar removal into a single catalytic filter [30–32]. As an example, an improved catalytic filter was fabricated by impregnating a nickel-based active ingredient into a porous ceramic candle filter, and tar removal at a gas velocity of 2–6 cm/s was investigated using benzene and/or naphthalene as model tar compounds [8,33–36]. Nacken et al. [31,32,37,38] studied two types of catalytic filters obtained by modifying bench-scale ceramic filters. Here, one of the filters was prepared by loading a nickel-based active ingredient on the pore walls of a porous ceramic candle by impregnation, and the other filter was a multilayer cylindrical ceramic candle comprised of, from the outer to inner layer, a filtering membrane, a filter candle, a free hollow cylindrical space, and a porous inner tube, where nickel-based catalyst particles were filled in the hollow cylindrical space to form a catalyst particle layer. Rapagnà et al. [39,40] placed the two types of catalytic filters in the bed freeboard of a fluidized bed gasifier, and obtained an overall increase in the gas yield and a decreased tar content of 58% and 79% for each respective filter. However, a fine particle layer adhered to the outer filtration surface of the ceramic candle, and the resulting pressure drop increased almost linearly with the progression of time. Rhyner et al. [18] conducted kinetics modeling of catalytic tar conversion. The simulation results were used to evaluate the different possibilities regarding three options integrating catalytic material into hot gas filter units with vertical and horizontal filter design. The integration options were the application of catalytic active filter elements [38], mounting an additional catalytic active foam cylinder at the inside of the filter element [31], and placing a monolith at the exit of the filter vessel. Placing a catalytic monolith or foam structure at the filter candle exit of a horizontal filter design was found to be unrealistic. Wang et al. [41] designed an auto-thermal reformer, with a porous ceramic tube circled by electronic wire, which was fixed inside an adiabatic reactor. NiO–MgO/γ-Al_2O_3 catalyst particles were loaded in the gap between the inside wall of the reactor and the outside wall of the porous ceramic tube. Clogging of the ceramic tube inside the reactor and the very large consumption of electrical energy were the two shortcomings of the auto-thermal reformer reactor.

According to the results reported by Mustonen et al., ceramic filters and granular bed filters are the most promising methods for hot gas cleanup [42]. Xiao et al. [43] analyzed the characteristics of various types of granular bed filters, and proposed that gran-