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Research article

Performance of biomorphic Silicon Carbide as particulate filter in diesel boilers

M. Pilar Orihuela [a, *], Aurora Gómez-Martín [b], José A. Becerra [a], Ricardo Chacartegui [a], Joaquín Ramírez-Rico [b]

[a] Dpto. Ingeniería Energética, Escuela Técnica Superior de Ingenieros, Universidad de Sevilla, Sevilla, Spain
[b] Dpto. Física de la Materia Condensada, Instituto de Ciencia de Materiales de Sevilla, Universidad de Sevilla, Consejo Superior de Investigaciones Científicas, Sevilla, Spain

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A B S T R A C T

Biomorphic Silicon Carbide (bioSiC) is a novel porous ceramic material with excellent mechanical and thermal properties. Previous studies have demonstrated that it may be a good candidate for its use as particle filter media of exhaust gases at medium or high temperature. In order to determine the filtration efficiency of biomorphic Silicon Carbide, and its adequacy as substrate for diesel particulate filters, different bioSiC-samples have been tested in the flue gases of a diesel boiler. For this purpose, an experimental facility to extract a fraction of the boiler exhaust flow and filter it under controlled conditions has been designed and built. Several filter samples with different microstructures, obtained from different precursors, have been tested in this bench. The experimental campaign was focused on the measurement of the number and size of particles before and after placing the samples. Results show that the initial efficiency of filters made from natural precursors is severely determined by the cutting direction and associated microstructure. In biomorphic Silicon Carbide derived from radially cut wood, the initial efficiency of the filter is higher than 95%. Nevertheless, when the cut of the wood is axial, the efficiency depends on the pore size and the permeability, reaching in some cases values in the range 70–90%. In this case, the presence of macropores in some of the samples reduces their efficiency as particle traps. In continuous operation, the accumulation of particles within the porous media leads to the formation of a soot cake, which improves the efficiency except in the case when extra-large pores exist. For all the samples, after a few operation cycles, capture efficiency was higher than 95%. These experimental results show the potential for developing filters for diesel boilers based on biomorphic Silicon Carbide.

1. Introduction

Diesel boilers are widely used in industry and domestic applications in order to heat or vaporize fluids. This fluid may be used in various processes or heating applications. Fossil fuels based boilers are a significant source of particulate matter emissions (Peckham, 2013). The presence of high concentrations of particulate matter in air is a significant environmental problem, not only due to the severe health problems it may cause (Prasad and Bella, 2010), but due to the associated economical cost (Zhang et al., 2008). Although small combustion plants (<50 MW) are not subject to specific standards in Europe yet ("Directive, 2001/80/EC," 2001), several Directives aimed at setting emission limits for the main pollutants have been already published. Directive 96/62/EC ("Directive 96/62/EC," 1996) published a list of pollutants and announced forthcoming limit values in the following years. Directive 1999/30/EC ("Directive, 1999/30/EC," 1999) set the first emission limits for the main pollutants in ambient air to be accomplished in average periods; among them, particulate matter below 10 μm in diameter (PM_{10}). Directive 2008/1/EC ("Directive, 2008/1/EC," 2008) established a list of activities that would be subject from then on to the granting of a permit for new installations; and included in the requirements for the permit granting the accomplishment of some general measures to avoid pollution. Directive 2008/50/EC ("Directive, 2008/50/EC," 2008) described common methods and criteria to assess the ambient air quality, and established reduction objectives and new emission thresholds for particulate matter, this time including also particles below 2.5 μm $PM_{2.5}$. In the United

* Corresponding author.
E-mail address: orihuelap@us.es (M.P. Orihuela).

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States, boilers and process heaters must comply with a specific regulation that sets limit values as a function of the fuel type, boiler size, use, and year of construction ("EPA 40 CFR," 2003).

The switch to biomass-fueled heaters implies a competitive advantage in the market and in the regulatory system, but does not prevent *per se* the release of solid particles. A study of Sippula et al. showed that the particle emission levels from wood-fired heating units could be higher than those of heavy fuel oil fired units (Sippula et al., 2009). In a later study, Sippula et al. determined that a wood-fired boiler might produce between 55 and 92 mg/MJ of fine particles (PM_1), and between 211 and 483 mg/MJ of particles in total (Sippula et al., 2007). Kaivosoja made a comparative study on the particle emissions from wood and oil boilers and observed that they had comparable PM_1 emission values. A wood wood-fired boiler might produce even more fine particles than a fuel oil-fired one if a filtration system was not used (Kaivosoja et al., 2013). To control solid emissions in industrial heaters one possible option is using conventional porous filters as physical barriers (Stoppiello et al., 2014).

Several authors have studied the characteristics and composition of particulate emission from boilers as a function of the fuel: A study of Fernandes et al. on the formation of fine particles in pellet-fired boilers revealed that the majority of the particles were smaller than 1 μm in diameter, and that the peak in the particle size distribution was between 50 and 130 nm (Fernandes and Costa, 2013). Huda et al. summarized the main findings obtained up to the date of their publication regarding the particulate emissions produced by boilers fired with agricultural waste (Huda et al., 2014). Limousy et al. provided with some particulate emission values for residential boiler fired with spent coffee grounds pellets (Limousy et al., 2013). Ozgen et al. (2012) measured the concentration and size distribution of the particles in the flue gases from different small boilers fired with different fuels. For light oil, Ozgen obtained an average concentration of $2 \cdot 10^6$ particles/cm^3 under hot sampling, with most likely particle size in the range 50–100 nm. Hays et al. (2008) presented similar results for a residential diesel boiler. They report an average geometric mean particle diameter of 36 nm, and provide with detailed physical and chemical information of these emissions.

When it comes to large industrial boilers, the use of baghouses (Sloat et al., 2016) and electrostatic precipitators (Caputo and Pelagagge, 1999) has been largely successful due to their high collection efficiencies, above 99% ("High-efficiency Particulate Filters for Industrial Boilers," 2010). In those cases, the removal of PM is usually performed jointly with the removal of other pollutants like SO_x or NO_x (Suk Kang et al., 2015). These filtration systems are, however, very complex and costly so their implementation becomes unfeasible in medium or small size facilities (Caputo and Pelagagge, 1999). For the abatement of particulate matter in small-scale boilers, other systems like multi-cyclones and core separators have been proposed (Hamilton et al., 2010). Small-scale wood boilers have been extensively studied. In this case, core separators and multi-cyclones are highly recommendable due to their efficiency and cost, however core separators are a newer technology and experience on wood-fired boilers is limited (Hinckley and Doshi, 2010).

To remove particles and other pollutants from the gas exhaust of coal-fueled boilers, Ha Tran et al. (2010) propose to use an Electrostatic Water Spraying Scrubber. The experiments they carried out produced collection efficiencies over 93% for all PM sizes. Patiño et al. designed and constructed a lab-scale tubular type Electrostatic Precipitator and tested it over a small-size diesel internal combustion engine (Patiño et al., 2016). Taking special care to prevent dirt accumulation on the arms of the star structure, they could reach collection efficiencies close to 100%. Catalysed wall-flow filters made of porous ceramic have also been proposed for particles removal in conventional small pellet boilers (Stoppiello et al., 2014). The study carried out by Stoppiello et al. reports abatement efficiencies for PM higher than 92%, but also several constraints like a high pressure drop, and the high working temperature needed to activate the catalyst.

Ceramic wall-flow filters are the most common way to abate solid emissions in diesel engines (Guan et al., 2015), and Silicon Carbide is, with Cordierite, one of the most popular materials (Adler, 2005) due to its high melting temperature, good heat transfer capability, low thermal expansion coefficient, excellent resistance to oxidation and corrosion, and high hardness and strength (Stobbe et al., 1993).

Biomorphic SiC (bioSiC) is a SiC-based porous ceramic material fabricated by Silicon melt infiltration of carbon preforms obtained from wood pyrolysis (Bautista et al., 2009), a low cost and environmentally friendly process in which a natural porous precursor is used as a template (de Arellano-López et al., 2005). The main advantage of this material with respect to other porous ceramics is its hierarchical microstructure, consequence of the preservation of the original wooden microstructure (Singh et al., 2003). Besides, the wide variety of existing plant species allows to almost tailor the morphology (density, porosity, pore size, etc.) of this porous microstructure (Kaul et al., 2006). BioSiC fabrication process has also several advantages compared to the fabrication techniques of traditional porous ceramics (de Arellano-López et al., 2005): lower processing temperatures, faster fabrication, additives are not necessary and complex shapes can be obtained easily by machining the carbon preform. Thus, by using this material we can improve some aspects of the use of SiC as a substrate for filters.

BioSiC has interesting characteristics for its application as diesel particulate filters that need to perform their task in aggressive environments. Some previous studies (Alonso-Fariñas et al., 2013) have demonstrated the feasibility of biomorphic SiC as filtration material for high temperature gasification processes. BioSiC permeability (Gómez-Martín et al., 2016a) and thermal conductivity (Gómez-Martín et al., 2016b) have been also recently characterized.

In this work, the performance of bioSiC as diesel particulate filter in boilers has been studied as a preliminary approach for identifying their interest and capacities for building advanced integrated particles abatement systems based on these materials. BioSiC is a low-cost and eco-friendly engineering ceramic material (de Arellano-López et al., 2005), the usefulness of which has already been demonstrated for several applications like catalyst support structures or bone implants (Singh et al., 2003). Through this work, a new application of this material has been analysed, while responding to increasingly severe pollution regulations, and to foreseeable future standards for diesel boilers. This paper presents, to the authors' knowledge, the first study about the use of bioSiC as substrate for diesel particulate filters, in this case for small diesel boilers. Despite the simplicity of the geometry of the samples tested in this work, the results obtained show their good performance and suitability for further developments. In addition, from the experimental results, an empirical correlation between filtration efficiency and permeability was developed.

For this purpose, an experimental set up has been developed for the integration of bioSiC filters, with different precursors and properties, in a real diesel boiler facility. The boiler, that works heating up thermal oil in a closed loop, doesn't operate constantly but through temperature regulated start-stop cycles as a function of the oil temperature. It is characterized by frequent start-stop cycles where diesel soot is generated. The results of the experimental campaign show the good performance of the samples, and the relevant differences in filtering efficiency depending on the wood