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Microwave-assisted regeneration of a perovskite coated ceramic monolith soot-trap

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ABSTRACT

Regeneration of a $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnO}_3$ perovskite coated ceramic monolith as a model soot filter, using carbon as model soot, has been performed in a travelling wave once-through microwave system. Thus, imitation of soot filter regeneration for diesel engines is attempted by carbon burn-off on a monolithic filter coated either with both perovskite and synthetic carbon or with carbon only. The results show that, in the presence of the perovskite, complete burn-off of carbon may occur. The energy consumption for regeneration is low, as compared to non-catalytic soot filter regeneration under the same reaction conditions. This work shows the viability of dielectrically sensitive oxidative catalytic coating materials for the proposed application.

Key words: microwave heating, carbon oxidation, perovskite, monolithic filter, in-situ filter regeneration and soot

INTRODUCTION

Diesel engines have an increasing market share in vehicle and generator markets due to their high efficiency and low fuel cost. The major drawback, however, is that pollution emitted by Diesel engines, particularly in the form of soot particulate matter, is of increasing health concern [1-3]. For this reason, European soot emission standards will be tightened in the coming 5 years [4]. Substantial efforts are devoted to developing new after treatment processes to match these emission standards. Techniques that have been designed to reduce the particulate emission include application of new fuel additives, modification of the engine structure, and the installation of exhaust-treatment systems [1, 5]. Among these techniques, a soot particle-trap filter has been considered as the best option. While a filtration efficiency of up to 90 % can be reached, an efficient way to carry out in-situ regeneration of these filter elements has not been found yet, the main challenge being the high ignition temperature of diesel soot [6-8].

In the last decade, microwave radiation has been discovered as an innovative tool for chemical reactions due to the enhancement of the reaction rate and selectivity in certain reactions [9-11]. The advantages of microwave heating include instantaneous and fast heating, easy control [6, 12]. Presently, we have been studying the feasibility of microwave heating as an alternative for in-situ soot filter regeneration. By periodically subjecting a soot filter coated with a microwave sensitive material to a dielectric field, soot ignition temperatures can be reached, which may subsequently allow soot filter self-sustained carbon burn-off.

The ideal coating material is also an active oxidation catalyst, allowing oxidation of formed side products during soot burn-off, such as CO. We chose La-Ce-Mn based perovskites as potential coating materials due to their favourable dielectric and

catalytic properties [13-17]. In this work, in-situ regeneration of a flow-through filter coated with synthetic carbon is carried out by microwave irradiation and compared to regeneration of a filter coated with both a perovskite and synthetic carbon. This reaction can be considered as a realistic model for regeneration of such a filter after exposure to Diesel soot.

EXPERIMENTAL

Material and preparation

A $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnO}_3$ perovskite was used as a coating material, manufactured by Praxair Specialty Ceramics using combustion spray pyrolysis. The material had a chemical purity of 99.9%. A ceramic cordierite monolithic substrate (Corning Inc.) was used for the preparation of laboratory samples (length = 40 mm, diameter = 15 mm, cell density = 400 cpsi). For soot combustion a synthetic carbon (Norit, RB2, 94 wt%, 6% ash) was used as 'model' soot. The dielectric properties of $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnO}_3$ perovskite has been measured by a dielectric probe (Agilent HP 85070C). These are compared to the properties of synthetic carbon and cordierite in table 1. For the perovskite/soot coated material, the monolith was first moistened with double distilled water, followed by coating with a perovskite suspension (60 wt%). After drying at room temperature, the monoliths were calcined at 1123 K, followed by a subsequent calcination on procedure at 1143 K, with heating at a rate of $2 \text{ K}\cdot\text{min}^{-1}$. Subsequently, coating with carbon suspension (13 wt%) was carried out, followed by drying at room temperature. The obtained monolith coded as monolith A contained 33 wt% perovskite and 3 wt% carbon. Another monolith was prepared by coating with carbon only, using the same procedure. This material (monolith B) contained 4.7 wt% carbon.

Microwave set up

The dielectric heating system is based on a travelling wave once-through set-up, with microwave radiation being absorbed by a water load positioned at the end of the wave-guide. The system consists of a microwave source (2.45 GHz, 1 kW), a circulator, a three-stub tuner section, a monomode microwave TE₁₀₀ cavity, and a water load, (figure 1). Reflection of microwaves is minimised by means of the stub tuners. The microwave source is protected from the reflected radiation by a circulator. The temperature in the sample bed is assessed by means of an optical fibre. A quartz sample tube (i.d. = 18 mm), is placed perpendicular to the direction of propagation, allowing the sample bed to be uniformly exposed to microwave radiation. The optical fibre is calibrated using thermocouples, by heating both an optical fibre and a thermocouple conventionally to 473 K, followed by stabilisation at 473 K for one hour. The lower detection limit of the optical fibre is 373 K.

Synthesised soot combustion

Temperature-programmed oxidation of the soot deposited filters with and without La_{0.8}Ce_{0.2}MnO₃ perovskite coating was performed in the single-mode cavity described above, using a O₂ (Praxair, 99.5%)/He (Praxair, 99.999%) gas mixture (v/v 5%/95%, GHSV = 5000 hr⁻¹). In each test the temperature was raised from 293 K to 1023 K at a maximum heating rate (295 K·min⁻¹) and maintained at the target temperature for one hour. The concentration of O₂, CO and CO₂ in the outlet was analysed by a mass spectrometry.

RESULTS AND DISCUSSION

The monolith coated with both perovskite and carbon (monolith A) was subjected to temperature-programmed oxidation by microwave heating. The background signal of CO, CO₂ and O₂ is subtracted from the obtained MS signal and corrected for the inlet pressure, using Ar as reference gas. The supplied incident power in terms of watts and the resulting temperature are shown in figure 2a, and the MS signal of the relevant components is in figure 2b. The maximum temperature of 1023 K is reached after 2.75 min and held there until the microwave was switched off. An incident power of about 200 W, of which about 85 % was absorbed, was needed to maintain this temperature. At a temperature of about 600 K the CO₂ concentration in the outlet gas starts to increase, while the O₂ concentration decreases. Maximum conversion towards CO₂ is reached after 2.5 min. Afterwards, the carbon oxidation rate decreases as a result of carbon burning off. No CO was detected in the outlet gas, indicating that carbon is converted completely to CO₂. When carbon is being burned-off, the perovskite coating, as a high dielectric loss material (see table 1), serves to keep the filter at a designed, sufficiently high, temperature during microwave heating. The carbon overall conversion is calculated by measuring the loss in weight after the microwave was switched off. We determined that about 100 wt% of the carbon has been burned off.

For comparison, the same temperature-programmed test was performed for the ceramic monolith coated with 4.7 wt% carbon only (monolith B). The results are shown in figure 3. Even by application of the maximum incident power (1 kW), the target temperature could not be reached (figure 3a). The maximum observed temperature was 960 K. Above 635 K, the appearance of CO and CO₂ are observed in the outlet gas, along with a drop of the O₂ concentration, indicating oxidation of carbon (figure 3b). Subsequently, the bulk temperature decreases to 680 K, and a corresponding decrease of the conversion rate for carbon oxidation is found. Our

results indicate that the oxidation rate and the temperature are a function of the amount of carbon present on the filter. As carbon is the only dielectrically sensitive component present in the material (see table 1), the heating efficiency (and thus the temperature reached) depends directly on the amount of carbon present. For low carbon contents, the microwave power is insufficient to reach the required temperature for carbon burn-off, and carbon does not oxidise any further. Our findings are in line with results from Palma et al. [8] for temperature-programmed soot regeneration of soot-loaded ceramic foam during microwave heating, who concluded that the discontinuation of soot oxidation is due to too low carbon contents. The carbon is not completely converted to CO₂, as evidenced by the appearance of CO. After the microwave was switched off, 57.3 wt% carbon had been converted, as determined from the loss of weight. This is substantially lower than the burn-off achieved by the use of perovskite as a coating material.

CONCLUSION

Microwave heating is shown to be a very fast and efficient technique for carbon oxidation. Regeneration of a monolith filter coated with La_{0.8}Ce_{0.2}MnO₃ perovskite shows complete carbon conversion at low energy consumption. However, during microwave-assisted regeneration of a monolith coated with only carbon, a considerable amount of CO is formed beside CO₂. The temperature and carbon conversion rate decrease, along with ongoing carbon oxidation. Dielectric heating of the remaining carbon is not sufficient to maintain the required carbon ignition temperature, leading to discontinuation of the carbon oxidation reaction. Our findings show the possibility to design a practical and effective microwave-assisted catalytic soot filter for Diesel engine, using perovskites as a coating material. The main

advantages are quick heating towards the soot ignition temperature, a high regeneration efficiency and low energy requirements. The high potential of the procedure requires further study of the design of microwave cavities and improvement of the soot filter with respect to thermal and chemical stability, in order to make this system commercially operational.

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Table 1: The dielectric properties, the dielectric constant (ϵ') and loss factor (ϵ'') of $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnO}_3$ perovskite and synthetic carbon.

	Dielectric constant (ϵ')	Dielectric loss factor (ϵ'')	Ref.
$\text{La}_{0.8}\text{Ce}_{0.2}\text{MnO}_3$	21.0	3.6	-
Synthetic carbon	~10	~3	[6]
cordierite	2.9	0.14	[6]

FIGURES

Figure1: Scheme of 2.45 GHz microwave set-up: (1) water trap, (2) oxygen trap, (3) mass flow controller, (4) microwave source (Muegge), (5) generator (Muegge), (6) circulator (Philips), (7) stub tuners (Muegge), (8) power sensor (Rhode & Schwarz), (9) optical fibre (Luxtron, accufiber), (10) quartz tubular reactor, (11) water load, (12) Quadrupole mass spectrometer (Balzers, Prisma QMS 2000).

Figure 2: Temperature and incident power (a), and O_2 , CO and CO_2 gas concentrations (b) as functions of time for a carbon and perovskite loaded ceramic monolith (monolith A), feed gas: O_2/He (v/v 5/95, GHSV = 5000 hr^{-1}).

Figure 3: Temperature and incident power (a), and O_2 , CO and CO_2 gas concentration (b) as a function of time for a carbon loaded ceramic monolith (monolith B), feed gas: O_2/He (v/v 5/95, GHSV = 5000 hr^{-1}).