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Research article

Impact of oxidizing honeycomb catalysts integrated in firewood stoves on emissions under real-life operating conditions

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ABSTRACT

Catalytic systems integrated in firewood stoves represent a secondary measure for emission reduction. This study evaluates the impact on emissions of two types of honeycomb catalysts integrated in different firewood stoves. The tests were conducted under real-life related testing conditions. The pressure drop induced by the catalyst's carrier geometry affects primary combustion conditions which can influence the emissions. A negative primary effect reduces the catalytic efficiency and has to be considered for developing catalyst integrated solutions. However, a significant net emission reduction was observed. The ceramic catalyst reduced CO emissions by 83%. The metallic catalyst reduced CO emissions by 93% which was significantly better compared to the ceramic catalyst. The net emission reduction of OGC (~30%) and PM (~20%) was similar for both types of catalysts. In most cases, the "Ecodesign" emission limit values, which will enter into force in 2022 for new stoves, were met although the ignition and preheating batches were respected. PM emission composition showed a lower share of elemental (EC) and organic carbon (OC) with integrated catalyst. However, no selectivity towards more reduction of EC or OC was observed. Further investigations should evaluate the long term stability under real-life operation in the field and the effect of the catalyst on polycyclic aromatic hydrocarbon (PAH) emissions.

1. Introduction

Worldwide > 2.7 billion people rely on firewood for heating and cooking [1]. In developing countries cooking is the predominate purpose for the use of firewood [2]. In Europe, manually operated firewood room heating appliances are classified in five different product groups: Firewood cookers (EN 12815) [3], inset appliances and open fireplaces (EN 13229) [4], roomheaters (EN 13240) [5], tiled stoves (EN 15544) [6] and slow heat release appliances (EN 15250) [7]. The most frequently used biomass based room heating appliances are firewood stoves according to EN 13240 standard [8]. Such stoves are commercially available as industrial end-user products or as pre- fabricated construction sets. They are installed as free-standing devices in residential homes and are connected to a chimney system. The nominal thermal heat output is typically in the range of 5 to 10 kW. In Europe, the stock of such stoves was estimated at around 25 million appliances which represent around 40% of the total stock of domestic room heating devices fired by solid fuels [8]. In Austria, the share of EN 13240 firewood stoves is also 40% with a total number of around 580,000 appliances [9]. The purchase prices for hand stoked firewood stoves range between 300 and 10,000 Euros (€). An average purchase price of 2000 € was reported for hand stoked firewood stoves [8].

Due to their contribution to harmful emissions, like PAHs [10,11] and particulate matter emissions (PM_{10}, $PM_{2.5}$) [12,13] they are regarded as critical technologies in terms of local air pollution [14,15,16]. In Europe, the residential sector contributes with 57% of total $PM_{2.5}$ pollution and is therefore the most relevant polluter of $PM_{2.5}$ emissions [1]. A high stock of old combustion systems [8,17] which performs worse compared to modern systems [18–20] and maloperation of users [21–23] are regarded as the most relevant factors for high emissions of carbon monoxide (CO), organic gaseous compounds (OGC) and particulate matter emissions (PM).

Consequently authorities are forced to implement effective measures in order to reduce emissions of combustion processes. At the beginning of the year 2022, firewood operated room heating appliances have to comply with specific emission limit values (ELV) which were

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Table 1

Chemical properties of used firewood and kindling material.

	Moisture content[a] (kg/kg)	Net calorific value (MJ/kg, d.b.)	Ash content (g/kg, d.b.)	Carbon C (kg/kg, d.b.)	Hydrogen H (kg/kg, d.b.)	Nitrogen N (kg/kg, d.b.)	Sulfur S (mg/kg, d.b.)	Chlorine Cl (mg/kg, d.b.)
Analysis standard	EN 14774-1:2009 [35]	EN 14775:2009 [36]	EN 14775:2009 [37]	EN 15104:2011 [38]			EN 15289:2011 [39]	
Beech firewood (“*Fagus sylvatica*”)	0.14–0.17	18.84	11	0.486	0.06036	< 0.001	93	36
Spruce kindling (“*Picea abies*”)	0.12	18.29	4.5	0.488	0.0611	< 0.001	77	43

d.b. = dry base.

[a] As received.

elaborated during the Ecodesign and Energy labeling process of the European Union. Accordingly, the maximum emissions measured during EN standard type testing for new stove technologies are 1500 mg/m^3 for CO, 120 mg/m^3 for OGC and 40 mg/m^3 for PM emissions (all ELV at STP conditions: 273.15 K/ 101,325 Pa, measured in the dry flue gas and referred to 13 vol% oxygen (O_2)) [24]. These ELV will set an equal requirements in Europe and it might be quite challenging for stove manufacturers to comply with these ELV.

In addition to the consequent application of primary measures for optimization of combustion conditions [25,26] the use of catalytic converters as secondary measure seems a promising technology towards low emission stove technologies [27–30].

For example, catalytic systems as retrofit applications could represent an option to decrease emissions from older stoves which are already in the marked and which will not be changed in near future (e.g. due to low family income). New stove technologies with integrated catalysts and combined primary optimization, e.g. by air staging and automatically controlled air supply, could represent an option to achieve a new state-of-the-art technology concerning low emissions and high efficiency in future.

Currently, catalytic systems are not commonly used in European stove technologies, neither as retrofit applications nor as integrated solutions. This is caused on one hand by the fact that national emission limit values have to be met only during official type testing which is possible without catalytic systems. On the other hand economic reasons and technological issues, like the risk of blocking or the long term stability, represent barriers for a widespread implementation of catalytic systems in stoves.

In a previous study combustion tests with two types (ceramic and metallic carrier) of integrated oxidizing honeycomb catalysts revealed a clear emission reduction potential on both, gaseous and particulate matter emissions [31]. Thereby the catalysts were integrated in a special test facility and the catalytic efficiency was determined under various conditions with different configurations of integrated catalysts. Depending on the space velocity the combustion tests showed catalytic conversion rates up to > 95% for CO, 65% for OGC and 35% for PM emissions.

This study aims at a validation of the catalytic efficiency of catalyst integrated solutions, as investigated by Reichert et al. [31] under specific test conditions. Therefore, the reductive effect of the two types of honeycomb catalysts integrated in different commercial firewood stoves on CO, OGC and PM emissions was assessed.

Furthermore, the impact of catalyst's carrier geometries on primary combustion conditions was quantified. The primary effect of catalyst integration results from an additional pressure drop in the post combustion chamber which influences volume flow of combustion air supply. Consequently, combustion conditions and combustion processes are affected resulting in different emissions. This study aimed at a quantification of the primary effect which is important to be considered for further development of catalyst integrated solutions and also when distinguishing between the exclusive catalytic emission conversion (catalytic coating) and the total effect of the catalytic system (catalyst's carrier geometry and catalytic coating) on emissions. For real-life emission impact the net emission reduction which represents the combination of primary effect and catalytic conversion is essential. In order to evaluate the real-life applicability and effectiveness as best as possible the stoves were tested by advanced test methods which aim at reflecting real-life operation different to the existing EN standard type testing method [32]. Therefore, transient combustion conditions, like the ignition and preheating by the first fuel batches were respected and included in the data evaluation.

A further aim was to collect data about the catalytic effect on the PM emission composition in order to identify a potential selectivity of PM reduction effect.

2. Material and methods

2.1. Fuel

Beech (“*Fagus sylvatica*”) firewood according to ÖNORM EN 14961-5:2011 standard [33] was used for all combustion tests (Table 1). The kindling material for the ignition batch was spruce (“*Picea abies*”). The firewood as well as kindling material was provided by a local firewood producer [34] as ready-to-use products. The firewood derived from trees grown in the Austrian province “Lower Austria” and was dried after splitting for at least two years under ambient conditions in covered piles. It was delivered in boxes with a volume of about 1m^3. The boxes were stored in an outside cabin until the combustion tests were carried out.

2.2. Oxidizing honeycomb catalysts

Two different types of honeycomb carriers (ceramic and metallic) were integrated in different firewood roomheaters. The honeycomb carriers were coated with a washcoat of aluminum oxide (Al_2O_3). The washcoat contained the catalytic active noble metals platinum (Pt) and palladium (Pd). Both types of catalysts (ceramic and metallic) were specifically developed for the use in manually operated firewood stoves. They are commercially available and are sold under the brand name “EnviCat® - Long Life Plus” by the company CLARIANT.

The catalyst based on a ceramic carrier material (cerhC) was round shaped with a diameter of 0.144 m and a cell density of 3.875 cells/cm^2. The metallic honeycomb catalyst (methC) based on a carrier of brazed stainless steel. The methC was also round shaped with a diameter of 0.149 m and a cell density of 7.750 cells/cm^2. The depth of both types of catalyst was 0.051 m. The shape of single cells of the cerhC was rectangular, the shape of single cells of the methC was trapezoid.

In this study also uncoated honeycomb carriers (“dummy”) were used. These dummies had no catalytic effect, but enabled an equal pressure drop and therefore equal primary combustion conditions. The physical data of these carriers were identical to the coated catalytic