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Catalytic efficiency of oxidizing honeycomb catalysts integrated in firewood stoves evaluated by a novel measuring methodology under real-life operating conditions

G. Reichert [a, b], C. Schmidl [a, *], W. Haslinger [a], H. Stressler [a], R. Sturmlechner [a], M. Schwabl [a], M. Wöhler [c], C. Hochenauer [b]

[a] BIOENERGY 2020+ GmbH, Inffeldgasse 21 b, 8010 Graz, Austria
[b] Graz University of Technology, Institute of Thermal Engineering – Thermal Energy Systems and Biomass, Inffeldgasse 25/B, 8010 Graz, Austria
[c] University of Applied Forest Sciences Rottenburg, Schadenweilerhof, 72108 Rottenburg a. N., Germany

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ABSTRACT

Catalytic systems integrated in firewood stoves represent a potential secondary measure for emission reduction. However, the evaluation of catalytic efficiency is challenging since measurements, especially for PM emissions, upstream an integrated catalyst are not possible. Therefore, a special test facility, called "DemoCat", was constructed which enabled parallel measurements in catalytically treated and untreated flue gas. The catalytic efficiency for CO, OGC and PM emissions was investigated under real-life operating conditions including ignition and preheating. The results confirmed a significant emission reduction potential (CO: > 95%, OGC: > 60%, PM: ~30%). The conversion rates of CO and OGC emissions correlated with the space velocity and the coated area of honeycomb carriers which represent key parameters for the integration design. A quick response of the catalytic effect of around 5–12 min after ignition was observed when reaching 250 °C flue gas temperature at the catalyst. Most effective CO and OGC emission conversion was evident during the start-up and burn-out phase of a firewood batch. This reveals an important synergy for primary optimization which focuses particularly on the stretched intermediate phase of a combustion batch. The catalytic effect on PM emissions, especially on chemical composition, needs further investigations.

1. Introduction

The use of wood in batch-wise operated firewood room heating appliances is the oldest and even most common way of woody biomass utilization [1,2]. Worldwide more than 2.7 billion people rely on wood for heating or cooking purposes [3]. The popularity of wood stoves in Europe is illustrated by a stock of around 65 million (Mio.) biomass room heating devices. The number of annual sold appliances in Europe is around 1 Mio [4]. In Austria, the stock of firewood room heating appliances was quantified at around 1.4 Mio [5]. They represent the most important source of providing renewable heat to residential buildings.

However, these appliances contribute significantly to local air pollution [6,7]. Especially in winter time, the use of wood in small-scale room heating appliances contributes significantly to PM10 and PM2.5 pollution [8,9]. Furthermore, carcinogenic and toxic emissions of gaseous and particulate carbonaceous components, like polycyclic aromatic hydrocarbons (PAH), tar and soot are emitted by such biomass room heating appliances [10,11]. These emissions have a negative effect on human health, e.g. causing respiratory problems, bronchial asthma and even premature death [12–14].

Consequently, authorities are forced to implement effective measures that enable and support an emission reduced utilization of wood in biomass room heating appliances.

In real-life operation the reasons for gaseous and particulate emissions of firewood operated room heating appliances are manifold. One important aspect might be the high stock of old appliances [4,15] which were found to have significantly higher emissions compared to modern types of appliances [16,17]. But, beside technological reasons, the user behavior and the operating conditions referred to the installation conditions, e.g. flue gas

* Corresponding author.
E-mail address: christoph.schmidl@bioenergy2020.eu (C. Schmidl).

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draught, can also influence the emissions and the energy efficiency performance [18–20].

Modern types of appliances are featured with implemented primary measures, like a well dimensioned combustion chamber design and an air staging concept. These primary measures aim at emission prevention by enabling optimal combustion conditions, especially during the intermediate phase of a combustion batch [21,22].

However, even modern and primary optimized stoves have combustion phases characterized by increased emissions, for example during the start-up and burn-out phase of a combustion batch or during the ignition batch [23,24]. Furthermore, modern types of firewood stoves are prone to a worse emission and efficiency performance when they are improperly operated, e.g. deviations to the suggested operation in the manual [18]. Unsuitable ignition procedure [25,26], incorrect fuel dimensions and fuel properties [27,28], an overloading of fuel [29] or incorrect adjusted air valve settings [30–32] result in increased emission levels. Additionally, different types of firewood species can result in different emissions and PM emission compositions [33,34]. Furthermore, thermal efficiency performance is strongly influenced by the operating conditions, especially by draught conditions [18,35].

Catalytic converters, in most cases used as retrofitted application [34,36,37], are feasible for an emission reduction. An advantage of catalysts in relation to other potential secondary devices, like filters or electrostatic precipitators (ESP), is the reductive effect on both, gaseous and particulate matter emission [38,39,40]. In addition, they operate also under non-optimal phases, e.g. during ignition (if the temperature is sufficiently high), during start-up and burn-out phases, or during critical operating conditions due to user behavior reasons [40]. In general, catalysts do not need electrical power supply. However, a certain temperature level is necessary to enable the catalytic process [40–42]. In some studies an external heating of the catalytic system was applied for that reason [37,43]. Many oxidizing catalysts work effectively when they are operated at temperatures of 300 °C–450 °C, especially for emission conversion of organic gaseous compounds (OGC) [41,42,44].

Without an external heating device this temperature level is usually not reachable with retrofit applications or only for a short time duration in wood stoves. Furthermore, the duration until the light-off temperature of the retrofitted catalyst is reached lasts around 20 min [38,40]. An alternative option would be to integrate the catalytic system in the combustion appliance where flue gas temperatures are sufficiently high and light-off temperatures of the catalyst are reached faster.

This study focused on an investigation of such approach. Therefore, the catalytic efficiency of two different types of platinum (Pt) and palladium (Pd) coated oxidizing honeycomb catalysts (ceramic and metallic carrier) was assessed as integrated solution. Since the evaluation of catalytic efficiency of integrated catalysts is difficult regarding to technical measurement reasons a novel measuring methodology was applied. Therefore, a special test facility, called "DemoCat", was constructed which enabled parallel measurement in catalytically treated and untreated flue gas. Using this novel test facility it was possible to evaluate the conversion rates of integrated catalysts regarding carbon monoxide (CO), organic gaseous compounds (OGC) and particle matter emissions (PM) under real-life operating conditions including ignition, preheating and several consecutive batches. Potential correlations between catalytic conversion rates of CO, OGC and PM emissions, space velocity and coated area of honeycomb carriers were investigated and analyzed. Furthermore, catalytic conversion rates of CO and OGC emissions during the characteristic firewood combustion phases, i.e. start-up, intermediate and burn-out phase, were assessed. The overall objective was to investigate the catalytic efficiency during different combustion conditions and to identify how integrated catalytic systems could be used as synergetic solutions for primary optimized firewood stoves. Finally, the study aimed at a profound knowledge of relevant criteria to develop and design catalyst integrated solutions.

2. Material and methods

2.1. Fuel

Beech ("*Fagus sylvatica*") and spruce ("*Picea abies*") firewood according to ÖNORM EN 14961-5:2011 standard [45] were used for all combustion tests (Table 1). The firewood and kindling material derived from trees grown in the Austrian province "Lower Austria". It was provided by a local firewood producer [46] as ready to use products. The firewood was stored covered outside until the respective combustion tests were carried out.

2.2. Oxidizing honeycomb catalysts

A noble metal catalyst – platinum (Pt) and palladium (Pd) based on a washcoat of aluminum oxide (Al_2O_3) – on two different types of honeycomb carriers (ceramic and metallic) was used in this study. Both types of honeycomb catalysts, called "*EnviCat® - Long Life*", were specifically developed and adapted for firewood combustion in manually operated stoves. They are commercially available in different shapes and dimensions. In total three different types of catalytic devices were used (Table 2). All catalysts were not used before the combustion tests of this study.

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 flow conditions. The physical data of these carriers were identical to the coated catalytic converters as it is given in Table 2. Catalysts and dummies were provided by the company CLARIANT.

2.3. DemoCat test facility

The DemoCat test facility was self-constructed and consisted of an adapted firewood stove and the subsequent measuring section (Fig. 1). The firewood stove was a roomheater classified according to EN 13240 [52] with a nominal thermal heat output of 10 kW. The round combustion chamber (diameter: 0.35 m, volume 0.0433 m^3) was lined with fire clay.

The DemoCat test facility enabled integrated testing of catalytic honeycomb catalysts. Therefore, the post combustion chamber of the firewood stove and the downstream flue gas measuring section was split into two symmetric parts. In one part a catalyst, in the other part a dummy of the same dimensions were integrated. This ensured equal pressure conditions in both measurement sections. The tightness of both sections was proved by applying a blower-door test with an overpressure of 10 Pa and measuring the leakage rate in both measuring sections. The leakage rate was below 1 m^3/h. According to pretests it was guaranteed that the total volume flow of the flue gas was equally distributed over both measuring sections. Due to the low flue gas velocities ($\leq$1 m/s) a continuous measurement, e.g. with a Prandtl or Pitot tube, was not possible. However, to identify potential deviations of volume flow conditions the static pressure drop of both measuring sections was continuously monitored.

The inner dimensions of the DemoCat box were 0.25 m × 0.25 m × 0.21 m (length × width × height). The part of the box for the integrated dummy and the catalyst were