Biomass and Bioenergy 105 (2017) 428–442

Contents lists available at ScienceDirect

Biomass and Bioenergy

journal homepage: http://www.elsevier.com/locate/biombioe

Research paper

Long term durability and safety aspects of oxidizing honeycomb catalysts integrated in firewood stoves

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

[a] BIOENERGY 2020+ GmbH, Inffeldgasse 21b, 8010 Graz, Austria
[b] Graz University of Technology, Institute of Thermal Engineering – Thermal Energy Systems and Biomass, Inffeldgasse 25/B, 8010 Graz, Austria

ARTICLE INFO

Article history:
Received 9 February 2017
Received in revised form
24 July 2017
Accepted 27 July 2017

Keywords:
Firewood combustion
User behavior
Long term durability
Honeycomb catalyst
Catalyst deactivation

ABSTRACT

Critical heating operating conditions, as emulated in the safety test series, showed that temperatures below 400 °C at the integrated catalysts result in deposited agglomerations on the flow cross-section area of the catalyst's surface and in the risk of increased pressure drops. The deposited material of safety tests consisted predominantly of carbonaceous components with a share of around 120 g kg^{-1} of OC and 280–450 g kg^{-1} of EC. The oxidation potential of deposited carbonaceous material by higher temperatures was confirmed by a minor share of EC and OC (<50 g kg^{-1}) on the catalyst's surface when a heating cycle with five batches was performed. Concluding a sufficient heating-up of catalyst integrated stoves is necessary to avoid deposition of carbonaceous agglomerations.

The long term tests resulted in deposited agglomerations of mineral particles on the catalyst's surface of both types of catalysts. The metallic honeycomb catalyst was more sensitive regarding blocking which was indicated by total blocked cells and a significant increase of pressure drop by 5.3 Pa. Due to the effect of agglomerated particles gaseous emissions increased significantly (CO around 300%, OGC around 45%) whereas PM emissions were reduced by 63%. The regeneration of catalyst performance was almost completely achieved by cleaning the catalyst with water and pressured air. For processing of blocking the open diameter of cells of the honeycomb catalysts play a relevant role. Therefore, in terms of real-life applicability the ceramic honeycomb catalyst seems to be more suitable compared to the metallic honeycomb catalyst.

1. Introduction

Firewood combustion in stoves is popular and widespread in Europe [1] and in addition an important technology for providing renewable heat to residential homes. Furthermore, the use of wood supports significantly the achievement of European CO_2 emission reduction targets [2] [3] [4]. However, old types of stoves emit high amounts of gaseous and particulate emissions [5] [6] [7] that negatively affect human health [8] [9]. Particulate emissions (PM10, PM2.5) and organic gaseous compounds (OGC), especially polycyclic aromatic hydrocarbons (PAH), are relevant emissions of firewood stoves [10] [11] [12] [13] causing respiratory health problems [14] [15] [16]. But, even advanced stoves incorporating primary optimization concepts, like air staging and well-designed combustion chambers [17] can emit considerable amounts of harmful gaseous and particulate emissions when they are operated under off-specification operating conditions [18] [19] [20] or by using firewood with inappropriate characteristics [21] [22]. Consequently, the industry is looking for solutions to optimize the combustion performance of firewood stoves regarding emissions and efficiency during regular use. Therefore, current research and development enhance the focus on the assessment of real-life stove performance using test procedures that reflect real-life operating conditions [23] [24].

The application of catalytic systems in firewood stoves as integrated or retrofitted applications is a well-known secondary measure to reduce emissions. For example, in the United States or in Canada, standard type test methods provide special limits and test procedures for stoves equipped with catalytic systems [25] [26]. In Europe, stoves equipped with catalytic systems are still not common, since the focus on technological development was set on the

---

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

http://dx.doi.org/10.1016/j.biombioe.2017.07.018
0961-9534/

optimization of primary combustion conditions [7] [17]. However, research and development is ongoing to combine primary and secondary measures in order to use the synergies of both optimization approaches. Further, the role of user behavior and its influence on operation performance is assessed and taken into consideration towards further development process [27] [28].

Catalytic systems are suitable to reduce gaseous [29] [30] and particulate [31] [32] emissions of firewood stoves, especially during the ignition process [33] and start-up and burn-out phases of a firewood batch [34] [35] where primary measures showed only limited effectiveness.

Honeycomb catalysts are commercially available and they are frequently used in the United States and Canada in firewood stoves [36] [37]. Typically, they are used with a bypass in order to avoid safety risks or malfunction due to blockage by deposited material. In previous studies a performance analysis regarding catalytic CO, OGC and PM emission reduction potential of a commercially available oxidizing honeycomb catalyst (CLARIANT EnviCat® LongLife Plus) was carried out [38] [39]. In different combustion tests the used type of ceramic honeycomb catalyst showed a reduction potential of 80–85% for CO, 40–50% for OGC and 12–55% for PM emissions.

In detail, the objective was to investigate and assess the risk of total blocking and subsequent impossible operation ability or operational problems due to particle agglomeration on the catalyst's surface. Furthermore, the potential decrease of catalytic conversion rate due to deactivation processes, e.g. thermal, chemical or mechanical deactivation, resulting by long term operation was assessed and evaluated.

2. Material & methods

2.1. Fuel

Beech ("*Fagus syslvatica*") firewood with an average length of 0.25 m according to ÖNORM EN 14961-5:2011 [40] was used for all combustion tests (Table 1). The firewood pieces and spruce kindling material ("*Picea abies*") were derived from trees grown in the Austrian Province "Lower Austria". Both, firewood and kindling material was bought as ready-to-use products from the local firewood producer HOFEGGER REINHARD (A-3250 Wieselburg). It was stored covered outside until the respective combustion tests were conducted.

2.2. Oxidizing honeycomb catalysts

Two different types of oxidizing honeycomb catalysts, both of the product line "EnviCat® - Long Life Plus", supplied by the company CLARIANT were used [46]. The first catalyst based on a ceramic carrier material with quadratic cells, the second catalyst based on a metallic carrier with trapezoid cells. The coating was equal for both types of catalysts and based on platinum (Pt) and palladium (Pd) on aluminum oxide (Al_2O_3). Two ceramic and two metallic catalysts were used in this study (Table 2).

2.3. Stoves

Two different firewood stoves (stove A: RIKA ECO RIKATRONIC^3, model number: 1300803 and stove B: WAMSLER SATURN S, model number: 10862) classified according to the standard EN 13240 [47] were used (Table 3). They are commercially available and represent commonly used stoves in terms of air staging and combustion chamber design. Stove A was a heavy stove providing heat storage stones on the top and at both sides outside of the combustion chamber whereas stove B was a light stove consisting of a steel envelope. Stove A was equipped with an automatic control device for combustion air supply. Hence, the combustion air supply is adapted by a control mechanism according to a temperature measurement in the combustion chamber. Thereby, the total combustion air flow is divided in two parts, primary air and secondary air. The automatic control system adjusts the primary and secondary air supply using an actuator connected to two dampers.

The combustion air supply of stove B was manually controlled by two dampers, one for primary air supply and the second for secondary air supply. Window flushing air of stove B was provided by two holes above the combustion chamber door. The amount of window flushing air is not controllable by the user.

The honeycomb catalysts were integrated in the post combustion chamber of both stoves (Fig. A1). Therefore, two mounts were used for stove A in order to clamp the catalyst just before the flue outlet. For stove B a small box was constructed directly downstream the original flue outlet and the honeycomb converter was placed in this box. Between the honeycomb converters and the steel body of the stove a heating resistant gasket material was used. For both stoves there was no bypass for the flue gas for the total heating operation times. The open diameter for the flue gas passing through the honeycomb catalysts was 13 cm (diameter of flow cross-section area). Consequently, the effective catalytic volume was 0.677 dm^3 for both types of honeycomb converters which was similar to the design of a previous study [35].

2.4. Test procedure, experimental set-up and measurements

The test approach was structured in two different combustion tests respecting the main objectives of this study as illustrated in Fig. 1.

2.4.1. Safety tests

For assessing the effect of critical heating operation, 20 single ignition batches were carried out under natural draught conditions for each type of honeycomb catalyst integrated in stove A and B (Fig. A1, Fig. 2).

Since only one batch per heating cycle was performed, the flue gas temperatures were comparatively low and the stove itself was not at steady state and still heating up. Consequently, potential agglomerations on the catalyst's surface are not completely burnt-

Table 1
Chemical properties of used firewood and kindling material.

Analysis standard	Moisture* (g kg^{-1})	Net calorific value (MJ kg^{-1}, d.b.)	Ash (g kg^{-1}, d.b.)	Carbon C (kg kg^{-1}, d.b.)	Hydrogen H (kg kg^{-1}, d.b.)	Nitrogen N (g kg^{-1}, d.b.)	Sulfur S (mg kg^{-1}, d.b.)	Chlorine Cl (mg kg^{-1}, d.b.)
	EN 14774-1:2009 [41]	EN 14775:2009 [42]	EN 14775:2009 [43]	EN 15104:2011 [44]			EN 1515289:2011 [45]	
Beech firewood ("*Fagus sylvatica*")	120–150	17.73	8.6	0.472	0.0616	<1.0	93	36
Spruce kindling ("*Picea abies*")	95	18.29	8.6	0.487	0.0631	<1.0	50	31

db. = dry base/*as received.