



Full Length Article

Kaolin as additive in wood pellet combustion with several mixtures of spruce and short-rotation-coppice willow and its influence on emissions and ashes

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ABSTRACT

In this study ten mixtures of spruce (*Picea abies*), short-rotation-coppice (SRC) willow (*Salix spec.*; Clone Tordis) and kaolin were pelletised and burned in a residential biomass boiler (12 kW). Focus is on usability of SRC willow in exchange of forestal biomass (spruce), as well as on kaolin as fuel additive for particulate emission reduction in small-scale biomass combustion. Moreover, fuel blends were intended to match quality criteria for the European market according EN 14961-2. Initially, ten raw material mixtures were pelletized and the produced blend pellets were physically and chemically characterized. Combustion experiments with these fuel blends show differences in gaseous emissions, chemical composition of emitted particles and boiler ash. Kaolin addition reduced emitted particle mass even in lowest kaolin concentrations (0.2 wt.%) in every combination of spruce and SRC-willow. Chemical analysis shows decreased K-contents in the collected particles for fuels with kaolin addition, but an increased share of Zn.

1. Introduction

The usage of solid biomass in combustion processes has increased as renewable energy source due to carbon neutral properties during the last decades [1,2]. Disadvantages of the increased usage of solid biomass are the additional need of forestal biomass accompanied by an increasing competition for high quality residues of wood industries. Material and energetic utilization made the expansion of raw materials increasingly important [3]. Moreover, the usage for energetic, respectively heating purposes by direct combustion can be identified as a major emission source of particulate matter (PM) [4,5]. Solid biomass combustion research and recent developments of new burners improved the carbon burnout and decreased organic particulate emissions mostly by primary measures (e.g. air-staging) and new shapes of fuels (e.g. wood pellets) [6,7]. Hence, the current focus is on reduction of inorganic particles, which originate from incombustible substances respectively the ash and are influenced of little account by the above mentioned technical options. Moreover, the mentioned increased usage is accompanied by a scarcity of forestal feedstocks and new solid biomass feedstocks from agriculture and agro-forestry have been discussed and tested recently for direct combustion purposes [8–10]. The main problem of alternative solid biomass feedstocks is their increased content of particle forming substances and differences in chemical composition of inorganic compounds compared to forestal feedstocks

[11,10,12]. Nevertheless, biomass from SRC is seen as a potential replacement of forestal biomass and have been evaluated before [13,14].

Raw materials used in this study are spruce saw dust and SRC-willow wood chips. The produced pellets should match A1 quality class according EN 14961-2 (ash content < 0.7 wt.%) to enable a consumer product, with exceptions for higher ash and additive contents for validation purposes within this study. Obviously, inorganic particulate emissions will become even more important at the combustion of fuels made from feedstocks with an increased amount of particle forming elements like the used SRC fuel blends.

The most important particle forming elements in solid biomass combustion are K, S, Cl and Zn. These substances are characterized by a temperature dependent release from the fuel bed to the gas-phase, chemical reactions in the gas-phase and a subsequent particle formation process [11,15–17]. There have been two approaches reducing inorganic particulate emissions from biomass combustion in the past: a reduced firebed temperature or fuel additives. Both approaches effectuate a retention of inorganic particle forming elements in the boiler ash. A reduced firebed temperature makes use of the release behaviour of inorganic substances and have been focused in recent works [18,19]. Fuel additives are used to influence the ash chemistry and capture particle forming elements in non-evaporating chemical structures remaining in the boiler ash [20–22]. Aluminosilicates have been identified to capture potassium (K) and Ca-containing additives capture S and

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Table 1

Recent publications on kaolin and limestone as fuel additive in solid biomass combustion.

Biomass	Kaolin	Limestone	
Oat grain	x	x	[20]
spruce and straw	x		[23]
pine bark, agricultural waste	x		[24]
willow, wheat straw and mixtures	x	x	[22]
miscanthus and tall fescue	x		[25]

Cl in the firebed during biomass combustion. Kaolin as aluminosilicate was used in the past as a natural clay material, as well as Limestone (CaCO_3). An overview of recent publications focusing this additives with several solid biomass fuels is listed in Table 1.

Kaolin as a mineral clay is composed mainly of Kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and can be used as fuel additive capturing particle forming K-compounds. The K-compounds KOH and KCl are captured by adsorption under reducing conditions in the fuel bed by Kaolinite [26]. Bäfver et al. [20] found a decreasing particle mass at kaolin addition in oat grain combustion, but an increase in particle number, as well as increasing gaseous HCl and SO_2 -emissions. Co-combustion of biomass with peat is also possible, because of a similar chemical composition as Kaolinite (high share of Si, Al, Fe, Ca [27,28]). Peat was used by several authors [25,27,28] as aluminosilicate additive and uses additionally the effect of Ca as sorbent of HCl and SO_2 [29,20]. Another Ca-containing additive is limestone (CaCO_3), binding Cl and SO_2 to CaCl and CaSO_4 , but can be influenced by a high share of Phosphor (P) [20,29]. Because of a high P-content in SRC raw material (500 mg kg^{-1}) compared to spruce raw material (21 mg kg^{-1}), Ca-containing additives were not considered in the present study to avoid reactions between P and Ca as described by Bäfver et al. [20] and kaolin as K-capturer was chosen. Moreover, detailed knowledge on the functionality of kaolin is available in literature, which describes the adsorption of K as an irreversible process [26,30]. Focus of the present work was on low shares of SRC-material to match the current availability for wood pellet producers in Europe. Adding mineral substances in solid biomass combustion results in the disadvantage of an increased ash content, because of their prevailing non-combustible properties. This aspect has to be kept in mind while using additives in combination with high-ash containing fuels in residential biomass combustion devices, which are usually intended for fuels matching EN 14961-2 with an ash content $< 0.7 \text{ wt.}\%$.

The objectives of this study are (i) the pelletization of several mixtures of spruce, SRC-willow and kaolin, as well as fuel characterization, (ii) the combustion in a residential biomass boiler focusing on combustion process and particulate emissions and (iii) chemical analysis of fuels, emitted particles and boiler ash.

2. Material and methods

2.1. Raw material

For wood pellet production two raw materials were used in different mixing ratios. Saw dust of unbarked Spruce (*Picea Abies*) was provided by a commercial pellet plant (FireStixx Holz-Energie GmbH) in March 2015, representing a common raw material in Central Europe for pellet production, as well as for residential heating purposes. Pellets from unbarked spruce fulfil EN 14961-2, respectively an ash content $< 0.7 \text{ wt.}\%$ easily. Raw material was already dried and the particle size was reduced by an industrial scale hammer mill to a suitable size distribution for pelletizing (Fig. 1). SRC raw material was obtained from a willow short rotation plantation (*Salix spec.*; Clone Tordis) located in Bad Schussenried in south-west Germany ($48^\circ 0' 44.12'' \text{ N}$, $9^\circ 39' 39.57'' \text{ E}$). Elevation of the growing area is 574 m above sea level, average annual temperature is 8.1° C and total annual average rainfall is 945 mm. The five years old trees (second rotation) were harvested by

a forage harvester and wood chips were transported to the pelletization lab. Subsequently, the wood chips were ground in a hammer mill (Muench SDHM 2) using a 6 mm screen and particle size distribution is compared with spruce saw dust in Fig. 1. The used SRC raw material represents a typical SRC plantation and harvesting in Central Europe, but at the moment it is only available in minor amounts for commercial pellet production. Wheat starch was used as binder which is common in commercial pellet production to support the densification process [31]. Water content of feedstocks was adjusted to 11 wt.% by adding water during the mixing process. In total, ten mixtures of raw materials with or without kaolin and a constant amount of binder were pelletized. An overview of the fuel blends is given in Table 3.

Raw materials have been analysed physically and chemically as described in Section 2.3 and results are shown in Table 2. Calorific value of SRC raw material is slightly decreased compared to spruce raw material. The ash content of SRC (1.42 wt.%) is considerably higher than the ash content of spruce raw material (0.26 wt.%) and chemical analysis of SRC raw material shows clearly increased values for N, S, Ca, K, Mg, P, Si and Zn compared to spruce raw material. The determined Cl contents are comparable between SRC (220 g kg^{-1}) and spruce (195 g kg^{-1}).

2.2. Pelletization

Pellets (6 mm in diameter) were produced using a semi-industrial pellet mill (Muench RMP 250) equipped with a rotating ring die and two fixed roller-cylinders. The ring die had channels with 6 mm diameter and 30 mm length. After start-up phase a constant equipment temperature was realized and pellets were produced with a constant feedstock mass flow of 100 kg h^{-1} . Feedstocks were weighed and mixed manually to match the aimed fuel blends according Table 3. Mixing ratios of kaolin were determined from the literature listed in Table 1 with at least one fuel blend matching an ash content $< 0.7 \text{ wt.}\%$. Pellets were screened after pelletization process using a vibrating screen (3.15 mm mesh) and cooled to ambient temperature by a counter stream cooler. Pelletization was done in one production run per fuel blend to ensure consistent conditions (Fig. 2).

2.3. Fuel and chemical analysis

Produced wood pellets and raw materials were characterized according EN 14961-2:2011 and are referenced in Table 4. Chemical analysis were carried out for raw materials, produced pellets, ashed pellets (ashed at 550° C , reused from ash content determination), boiler ash and particulate matter. Used methods and standards are listed in Table 5. Three samples of each raw material and each fuel blend were sampled and pulverised to 1 mm with a cutting mill (Fritsch Pulverisette 19). These samples were used for the subsequent sample preparation. Before wet chemical analysis with Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and Photometer a microwave digestion was done with 2 ml HF, 10 ml HNO_3 and 10 ml H_3BO_3 for 0.1 g samples. A combustion digestion was used before Ion Chromatography (IC) Analysis for Cl determination. XRF measurements were done with four particle loaded filters per fuel blend and Si is not charted, because of its origin from used quartz-fibre filters. Moreover, five filters were analysed unloaded, but thermally treated as described before and measured as blank tests.

2.4. Combustion experiments

The residential pellet boiler (KWB, Easyfire 12 kW, year of manufacture 2012) was equipped with an underfeed-burner. Rotational speed of the stoker was manually controlled, aiming an average thermal output of 11 kW at full load. The chimney draught was set to $12 \pm 2 \text{ Pa}$ and the boiler was running $\geq 90 \text{ min}$ at full load prior to starting measurements in order to achieve steady-state conditions. The calculated

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