Fuel 232 (2018) 384–394

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Content of potassium and other aerosol forming elements in commercially available wood pellet batches

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A B S T R A C T

In wood fuels, potassium is by far the most abundant aerosol forming component. Given this predominant role, a major contribution for meeting the tightened total particulate matter emission thresholds on European level could thus be expected by the introduction of an upper limit for potassium content in wood pellets. To evaluate options to introduce such an upper limit for the potassium content of wood pellets, an extensive pellet screening was performed in Germany covering 22 of the 46 EN*plus* certified wood pellet producers representing more than 50% of the current German wood pellet production of about 2 million t/year. This screening was accompanied by the compilation of wood pellet fuel data from different sources. In total, analysis data of 249 high quality wood pellet samples has been evaluated. Almost all pellet samples originated from European production whereof the majority were pellets from certified German pellet producers. The pellet screening revealed a very large variation in the potassium content of high quality wood pellets ranging from below detection limit to well above 1000 mg/kg d.b. The results also confirmed the predominant role of potassium among the aerosol forming elements potassium, sodium, lead and zinc in wood samples. Furthermore, the screening highlighted that the ash content is not sufficient to evaluate the potassium content of wood pellets. Analysis of selected raw materials from three different pellet producers revealed a strikingly large variation in the potassium content of the raw materials. Consequently, controlled adjustment of the potassium content in wood pellets in the production process would require dedicated mixtures of different raw materials and the limitation of the share of raw materials with particularly high potassium contents. In this way, the potassium content of wood pellets could be limited and this could contribute to the compliance with strict particulate emission thresholds for small scale combustion appliances. Prerequisite for such an approach would be a fast, simple and cheap possibility to determine the potassium content of raw materials for wood pellet production at the production sites and the incorporation of the potassium control in the quality assurance of the pellet production process. Consequently, wood pellets could be produced that facilitate compliance with particulate matter emission threshold without secondary emission reduction measures.

Abbreviations: A, ash content; aq, aqueous; a.r, as received; BImSchV, Bundesimmissionsschutzverordnung (Ordinance of the German Federal Immission Control Act); BMEL, Bundesministerium für Ernährung und Landwirtschaft (Federal Ministry of Food and Agriculture); d.b, dry basis; DBFZ, Deutsches Biomasseforschungszentrum gemeinnützige GmbH; DIN, Deutsches Institut für Normung (German Institute for Standardization); DT, deformation temperature; DV, variation over one production day; EN, Europäische Norm (European Standard); FT, flow temperature; FP7, EU's Seventh Framework Program for Research; HT, hemisphere temperature; HoPeS, Holzpelletscreening (wood pellet screening); ICP-MS, inductive coupled plasma mass spectroscopy; ICP-OES, inductive coupled plasma optical emission spectroscopy; ISO, International organization for standardization; M, moisture content; n, number; n.m., not measured; NREAP, National Renewable Energy Action Plan; P_i, pellet sample; PM, particulate matter; PVC, poly vinyl chloride; Q, net calorific value; R^2, coefficient of determination; RM, raw material; SST, start shrinking temperature; STP, standard temperature and pressure; TPM, total particulate matter; WV, variation over several consecutive production days of one production week

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https://doi.org/10.1016/j.fuel.2018.06.001
Received 7 August 2017; Received in revised form 16 May 2018; Accepted 1 June 2018

1. Introduction

Solid biomass is considered as the most important bioenergy source in order to achieve the renewable energy targets for 2020 outlined in the National Renewable Energy Action Plans (NREAPs) of the European member states [1]. It is estimated to represent about 38% (3.9 EJ) of the anticipated 20% share of renewable energy sources (10.3 EJ) in the EU gross final energy consumption by 2020 and will be with 3.4 EJ (72%) the dominating energy source for renewable heating [2,3]. Especially for residential heating, utilization of solid biomass like firewood, wood chips or wood pellets is quite popular and widely used. Application of wood pellets in automated boilers provides several advantages such as low emission levels and a high customer convenience due to fuel quality standards [4] and established fuel certification systems like EN*plus* [5]. However, biomass combustion, in particular in small scale heating appliances < 100 kW, is also recognized as one of the main sources for anthropogenic fine dust pollution which is known to have adverse effects on the pulmonary and cardiovascular system [6–9]. Consequently, in order to avoid a negative impact of biomass combustion on air quality and human health, emission thresholds have been established on national level. The First Ordinance of the German Federal Immission Control Act (1. BImSchV) for residential heat appliances currently sets ambitious total particulate emission (TPM) thresholds which are the strictest thresholds within Europe and North America for this size of boilers [10,11]. During type testing which is required for introducing a product into the market and during the compulsory biannual inspection measurements for boilers, TPM emission levels of $\leq 0.02\ g/m^3$ (STP, 13 vol% O_2) have to be met [10]. On European level, similarly tightened thresholds of $\leq 40\ mg/m^3$ TPM (STP, 10 vol% O_2) corresponding to about $0{,}03\ g/m^3$ (STP, 13 vol% O_2) are included in the European Ecodesign Directive for solid fuel boilers (Directive 2009/125/EC, LOT15), which have to be met as of 1st January 2020 [12]. Reliable compliance with these requirements may be supported by secondary emission reduction measures like dust separators. Under the current frameworks, however, these are often not economically feasible as add-on solutions for small scale wood combustion appliances and thus the currently effective TPM thresholds pose a challenge for wood boiler manufacturers and operators.

In modern, highly automated stoves and boilers complete combustion is commonly achieved resulting in particle emissions that originate predominantly from the ash forming elements of the fuel and consist mainly of inorganic material [13–17]. Typically, particle emissions contain a fine fraction with particles < 1 µm (i.e. aerosols, PM1) which makes up about 90% of the TPM [15,18,19]. The remaining coarse fraction is usually composed of non-volatized minerals such as alkaline earth metal species (Ca, Mg) as well as some Si, P and Mn which are entrained into the flue gas [20]. In contrast, the fine particle fraction is dominated by alkali (K, Na) metal chlorides and sulfates and may also contain further volatile ash components like Zn and Pb [15,17,21]. The fine particles are believed to be formed by vaporization of volatile ash forming elements during the combustion process and subsequent condensation and nucleation upon flue gas cooling [13,22,23]. In particular, soluble K compounds are expected to be easily vaporized [24–26]. This assumption is supported by the observed reduction of K release and diminished TPM emission during combustion of leached biomass samples [27–33]. In wood fuels, K is by far the most abundant aerosol forming component. During combustion experiments with different wood fuels, higher amounts of K typically resulted in higher TPM emission levels [14,34].

However, only part of the aerosol forming matter is released to the gas phase while a substantial share is retained in the bottom ash [22,23,35,36]. The released amount and the actual release mechanism depend on a number of factors, e.g. fuel ash composition and fuel bed temperature which in turn is dependent on parameters like boiler geometry, combustion technology, air/fuel ratio, boiler load etc. [13–16,37–40]. While chlorine generally facilitates K release [22,24,41,42], some elements may also support the retention of K in the bottom ash. During their studies with various annual biomass types Knudsen et al. showed that a significant difference in the K release can be found between Si lean fuels ($K/Si \gg 1$ mol/mol) and fuels with a significant Si content for which a substantial K retention was observed [22]. Studies with wheat straw and rice straw-wood mixtures indicated that K can be incorporated into the silicate structures hampering volatilization [40,43]. However, the work of Knudsen et al. also highlighted that based on the high affinity of K for Cl [25,40] significant fractions of K can still be released from Si rich fuels if they also contain large amounts of Cl. Furthermore, alkaline earth metals are known to compete for network positions in the silicate network [43]. Thus, vaporization of K in Si rich fuels is facilitated in case of high molar (Ca + Mg)/Si ratios > 2 mol/mol [22].

Despite the complex mechanism and various interactions involved in K release and particulate matter formation, strong correlations have been reported by various authors between the PM1 emission levels and the content of (i) K + Na + Pb + Zn [44], (ii) K + Na + Cl + S [14,34], (iii) K content [34] as well as between the level of TPM emissions and the K content [45]. However, data for fuels with low K content (i.e. typically for woody biomass) is rather limited and scattering increases with decreasing K content [46].

Given the predominant role of K as an aerosol forming element in wood fuels, a major contribution for meeting the tightened TPM emission thresholds on European level could be expected by the introduction of an upper limit for K content in wood pellets. Based on the available literature on particulate matter emissions from pellet combustion [17,45,47–56] as well as emission measurements performed during wood pellet combustion experiments at DBFZ [55,57], a critical K content of about 500–600 mg/kg d.b. could be identified. Combustion of wood pellets with K levels significantly above this critical content is likely to cause TPM emissions above $0.02\ g/m^3$ (STP, 13 vol% O_2) during full load boiler operation. Investigations of the chemical composition of wood fuels which have been performed both concerning differences between selected wood species as well as within one wood species revealed substantial differences in the content of aerosol forming matter [58–60]. To some extent, higher potassium contents in wood fuels could be traced back to contamination with bark and needles which are naturally containing higher amounts of K or with soil [61]. Chandrasekaran et al. studied various wood pellet samples during a wood fuel screening in the United States [60]. In total, 132 wood pellet samples were analyzed. None of the investigated pellet producers was certified and various samples did not meet the wood pellet quality standards set by DIN EN ISO 17225-2:2014, class A1 which is the wood pellet quality most commonly used in small scale combustion appliances for residential heating. In particular, the ash content was significantly higher than required by the quality standard in various cases. It is thus difficult to derive conclusions for the German and European pellet market that is dominated by high quality and commonly certified wood pellets for small scale combustion appliances < 100 kW [62]. The results revealed nevertheless the wide variety in the wood fuel composition as well as the requirement for standardization. Duca et al. analyzed the quality of 130 commercially available bagged wood pellet batches obtained in Italy covering both certified and uncertified pellets [63]. However, the focus of this study was the compliance with the wood pellet standard EN 14961-2 (superseded by DIN EN ISO 17225-2:2014) and thus only very limited information concerning the elemental composition of the tested pellet batches was provided. Currently, only the elements N, Cl, S, as well as the heavy metals As, Cd, Cr, Cu, Hg, Pb and Zn are included in the wood pellet quality standards set by DIN EN ISO 17225-2:2014, class A1 while potassium and sodium are not included.

In order to evaluate the options to incorporate K in wood pellet standardization the study aims at:

(i) estimating the share of pellets with elevated K content above