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Determination of off-gassing and self-heating potential of wood pellets – Method comparison and correlation analysis

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ABSTRACT

Several methods for identifying the phenomena of self-heating and off-gassing during production, transportation and storage of wood pellets have been developed in recent years. Research focused on the exploration of the underlying mechanisms, influencing factors or the quantification of self-heating or off-gassing tendencies. The present study aims at identifying a clear correlation between self-heating and off-gassing. Thus, different methods for determining self-heating and off-gassing potentials of wood pellets are compared. Therefore, eleven wood pellet batches from the European market were analyzed. For this investigation, three methods for the determination of self-heating, like isothermal calorimetry, oxi-press and thermogravimetric analysis, and four methods for off-gassing, like volatile organic compound (VOC) emissions measurements, gas phase analysis of stored pellets in a closed container by offline and by glass flask method and determination of fatty and resin acids content, were performed. Results were ranked according to the self-heating and off-gassing tendency providing a common overview of the analyzed pellets batches. Relations between different methods were investigated by Spearman's correlation coefficient. Evaluation of the results revealed an equal suitability of offline and glass flask methods to predict off-gassing tendency and indicated a very significant correlation with isothermal calorimetry for the identification of self-heating tendency. The thermogravimetric analysis as well as the fatty and resin acids determination proved to be insufficient for the exclusive assessment of self-heating and off-gassing tendency, respectively.

1. Introduction

Self-heating and off-gassing of several gases occur during storage and transportation of wood pellets. Both phenomena pose a significant health risk as well as financial risk to producers, operators and customers along the wood pellets supply chain. Larsson et al. [1] documented examples of eight fires caused by self-heating and spontaneous ignition during the storage of wood pellets that happened during 2004 to 2012. Furthermore, off-gassing emissions from wood pellets led to at least nine reported fatalities connected to the transportation or storage of wood pellets since 2002 [2].

During storage of solid biofuels, the formation of heat can be observed. In fact, detailed descriptions of several incidents of fires in wood pellet silos were reported by Persson et al. [3]. Oxidation of wood extractives is considered to be a dominant source of heat responsible for self-heating of biomass aided by various other exothermic processes such as biological processes or adsorption-desorption of water caused by the differences between real and equilibrium moisture concentration in storages [4,5].

Several different methods are proposed to determine the self-heating and self-ignition risk of biomass samples [6,7]. Most of the testing methods have derived from coal analyses. Various biomass fuels have been investigated by these methods, however, wood pellets have not been frequently examined [8]. Several studies analyzed self-heating

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properties of different biomass fuels employing isothermal calorimetry [9,10], isothermal oven procedure [11], and thermogravimetric analysis (TGA) [8,12–14].

In addition to the self-heating phenomenon, wood pellets are known to emit various gases during transportation and storage. Since 2004, investigations have indicated the formation of carbon monoxide (CO), carbon dioxide (CO_2), different volatile organic compounds (VOCs), methane (CH_4) and hydrogen (H_2) [15–19]. Simultaneously, oxygen depletion in the surrounding air was observed [20,21]. The mechanisms responsible for the formation of emissions have not been completely clarified yet. The first assumption about the causative reactions was that oxidation of wood extractives like fatty acids are the cause for the formation of CO [22]. Likewise, a further study indicated that fatty acids and resins have an important role on off-gassing [23]. A recent study confirmed the important role of wood extractives and identified the mechanistic pathway of CO formation. According to Rahman and Hopke, the reaction was initiated by the autoxidation of unsaturated wood extractives. The autoxidation of fatty acids and terpenes causes the formation of hydroxyl radicals. These react with the hemicellulose leading to the release of CO. However, the authors mentioned that this mechanism was unlikely to be the cause of the high CO emissions in reported fatal accidents [24].

In literature, different methods for the determination of off-gassing are described [16,19,23,25–27]. All these methods have in common that a defined amount of pellets is stored in a closed container with a specified volume for a defined period. A further similarity between these methods is that gas components in the headspace (the gas phase above and between the stored pellets) are measured. The previously used off-gassing methods vary concerning the amount of stored pellets, the prevailing temperature, the volume of headspace and the intervals of gas measurements, respectively. Moreover, gastight or ventilated storage systems are used. During storage of pellets, determination of wood extractives and its relevance for off-gassing was investigated trying to correlate the off-gassing with either single fatty acids or the total amount of extractives [16,28,29].

Although the underling mechanisms and degree of self-heating of wood pellets as well as off-gassing of CO, CO_2, CH_4 and VOCs from pellets during storage and transportation are not fully understood, there exist some clear indications that the two phenomena are connected since both assume the oxidation of wood extractives as main cause. Efforts have already been made to investigate a potential relation between off-gassing and self-heating. Two studies investigated the relation between self-heating and the content of fatty acids as well as the release of VOCs and a possible relation to the content of fatty acids. Both studies revealed that besides autoxidation processes of fatty acids and resins in sawdust and wood pellets during storage, the temperature within storage piles could increase up to 60 °C. The same studies reported a reduction of the content of fatty and resin acids with simultaneously decreasing emissions of aldehydes and ketones [16,29]. Therefore, the phenomena off-gassing and self-heating of wood pellets are believed to be closely connected.

The objective of the present work is to examine a potential correlation between off-gassing and self-heating occurring in wood pellets during storage. In order to reveal such a relation, eleven wood pellets batches are investigated using seven methods analyzing:

(i) the self-heating tendency utilizing isothermal calorimetry, oxi-press, TGA and
(ii) the off-gassing tendency employing measurements of the amount of emitted VOCs (specifically aldehydes and ketones), as well as the determination of formed CO, CO_2 and partially CH_4 in the gas phase above the pellets with two methods (offline and glass flask methods) and determination of fatty and resin acids content. Fatty and resin acids in pellets indicated the degree of autoxidation and were associated to off-gassing emissions.

Table 1
Investigated wood pellets batches.

Pellets batch number	Pine/spruce ratio	Production site	Diameter in mm
1	100/0	DBFZ	6
2	0/100	DBFZ	6
3	100/0	German pellets producer 1	6
4	20/80	Swedish pellets producer 1	8
5	0/100	German pellets producer 2	6
6	40/60	Swedish pellets producer 2	6
7	60/40	Swedish pellets producer 3	8
8	50/50	Swedish pellets producer 4	8
9	100/0	BIOENERGY 2020+ GmbH	6
10	100/0	DBFZ	6
11	20/80	German pellets producer 3	6

The results were ranked regarding their total self-heating and off-gassing tendency. Moreover, the results from seven methods were compared with each other using Spearman's correlation coefficient [30]. Thus, the comparison of the results from the individual methods shall provide insight into the question whether pellets, which show a high off-gassing reactivity, also exhibit a high self-heating reactivity.

2. Material and methods

2.1. Wood pellets batches

The self-heating and off-gassing behaviour of wood pellets was studied using eleven wood pellets batches (Table 1). The pellets were investigated in the course of the European research project SafePellets [31]. The initial SafePellets project sample numbers are listed in Table 1 in the Supplementary material since the pellets were previously studied in Larsson et al. [1]. The pellets originated either from commercial German and Swedish pellets producers or from lab scale production. The wood pellets batches were selected to represent the most commercially available pellets on the European market [32]. Therefore, pellets made of *Picea abies* (spruce), *Pinus sylvestris* (pine) or mixtures of both were investigated. The ratio was realized by mixing the raw material before pelletizing. All pellets produced at the commercial sites were packed by the pellets producers in 15 kg bags immediately after production and sent to Deutsches Biomasseforschungszentrum gemeinnützige GmbH (DBFZ). It was crucial to ensure that all participating research institutes would perform their studies on pellets with equal characteristics. Thus, to homogenize, the total amount of each batch was combined on a clean plastic sheet and manually mixed thoroughly for 10 min. Afterwards, 5 kg pellets were immediately packed in air tight plastic bags and sealed. The used type of bags did not emit any volatiles that could interfere with the performed experiments. The pellets were sent to the research institutes and stored at −18 °C until analysis.

2.2. General characterization – determination of solid biofuel characteristics

All batches were analysed according to the requirements of standard EN 14961-2 for solid biofuels which was recently superseded by ISO 17225-2, i.e. fines content, elementary analysis (carbon, hydrogen and nitrogen content), moisture content, ash content, total content of sulphur and chlorine, net calorific value, major and minor elements [33,34].

2.3. Determination of the self-heating tendency

Three different methods were used to evaluate the tendency of the eleven wood pellets batches for self-heating, i.e. isothermal calorimetry, oxi-press and TGA.

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