



Assessment of self-ignition risks of solid biofuels by thermal analysis



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HIGHLIGHTS

- Flammability data of biomasses are presented showing risks of self-ignition.
- Ground type for sampling preparation has no influence on flammability of biomasses.
- Lignocellulose composition influences the self-ignition tendency of biomasses.
- Activation energy is not a discriminant variable for the self-ignition of biomasses.
- Oxidation temperature characterised the self-ignition risk of biomasses.

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ABSTRACT

Storage of various types of biomass can result in spontaneous combustion processes, as they are capable of absorbing oxygen to produce exothermic oxidation reactions. Self-combustion is frequent in the storage and handling of biomass products and there are several factors related to their physical and chemical properties that influence their thermal susceptibility, that is, their tendency to oxidise and the subsequent ignition of the substance.

Biomass from agriculture, forestry and waste origins with different chemical compositions are studied to analyse their behaviour and to determine the main factors that have an influence on the self-ignition risk. Among those factors, chemical composition, physical treatments and flammability characteristics are studied. Also thermal analysis methods are included in this study. Based on classical techniques of thermogravimetry and differential scanning calorimetry, particular experimental applications are used to assess the self-ignition risk of biomass products. Results are statistically analysed looking for correlations and grouping of samples. Statistical analysis concludes that the chemical composition of the biomass has an overriding role in characterising the self-ignition tendency of these biomasses.

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1. Introduction

The main source of increasing carbon dioxide concentration in the atmosphere is the burning of fossil fuels like oil and coal, along with deforestation of tropical forests [1]. According to the World Meteorological Organization [2], the amount of greenhouse gases in the atmosphere is reaching a higher record every year. Between 1990 and 2012, a 32% increase in radiative forcing, (the warming effect on our climate due to carbon dioxide and other long-lived gases that trap heat, such as methane and nitrous oxide), was recorded.

Since the beginning of the industrial era in 1750, the global average concentration of CO₂ in the atmosphere has increased by 41%, methane by 160% and nitrous oxide by 20%. Only about half of the CO₂ emitted by human activities remains in the atmosphere, with the rest being absorbed in the biosphere and in the oceans.

The US National Oceanic and Atmospheric Administration [3] reported that the barrier of 400 ppm measured of CO₂ in the atmosphere was exceeded for the first time in 2013.

The World Energy Investment Outlook [4] estimates that the world would need to increase crude oil production by the equivalent of four Saudi Arabia's production by 2030 in order to meet decreasing production and rising demand. This, however, is impossible and unconventional fuels cannot fill such a deep gap.

The almost exclusive dependence on crude oil is evidently not ideal. Real or predicted alterations of crude oil supply have

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previously led to strong increases in crude oil prices and led to economic insecurity. For that reason, diversifications of primary energies for fuel production will be necessary, with to renewable forms of energy being of particular interest.

Renewable energy has become more significant in the world energy market and will play a very important role in the longer term. It makes up an important part of the solution by reducing import dependency, diversifying sources of production, and contributing to sustainable growth in Europe [5].

The Intergovernmental Panel on climate change [6] established that by 2100 the participation rate of biomass in the energy world's production must be between 25% and 46%. Although the availability of biomass is abundant in Europe, the supply is not yet organized in many cases. The promotion of a true European market for biomass is needed. A solid biofuels pioneer in this European market comes from local forest industry or waste product of maintenance and tending activities in forest industry. In some European countries this market has grown rapidly in recent times. In addition, production of high-density pellets allowing their transportation over long distances, has considerably improved the situation.

All such solid biofuels are capable of reacting with oxygen in the air, as is the case of agricultural materials, whose self-ignition phenomena are well known [7]. At room temperature, the oxidation reaction tends to be extremely slow, but is exothermic, meaning it can cause an increase in the temperature of the material if the generated heat is not dissipated fast enough [8]. These reactions mainly depend on the moisture of the sample, since it is caused by microbiological heating and fermentation [9]. It has been observed at around 30–50° Celsius in several solid biofuels but it is not detected by conventional thermogravimetric analysis [10,11].

If the heat produced in these reactions is not dispersed outward, the temperature will increase, causing an increase in the rate of the exothermic oxidation reaction, generating heat faster. This will produce a greater increase in the temperature, which will further increase the reaction rate so that the process is no longer stationary, it will lead to overflow of heat balance, resulting in a self-heating of the organic material which can lead to decomposition and ignition [12,13].

Chemical composition of several biomass products have been extensively studied, including traditional and complete proximate, ultimate and ash analyses, finding out that biomass composition is significantly different from that of coal and that variability among biomass composition is greater than for coal [14]. The burning characteristics of these biofuels affect the capacity to undergo the self-ignition process [15] and often relate to some of the characteristic parameters of the flammability of solids, as their ignition temperatures [16].

Standard Thermogravimetry (TG) analysis is a technique widely used in order to study the decomposition reactions of solids by heating [15,17,18].

These techniques have been used to detect the temperatures at which the exothermic processes start for solid products of natural origin, such as fossil fuels, biomass or food products of different origin, and to estimate kinetic characteristics of the oxidation process [19]. Comparing these temperatures between different products is possible to establish a relative scale of the tendency to spontaneous combustion, since the lower the temperatures at which the exothermic processes are initiated, then the greater the tendency of the product to self-ignite [20]. This procedure has been used for different types of coals [21], for some food products [7] and sewage sludge [22], obtaining satisfactory results.

However, it has been detected a lack of detailed data on thermal analysis techniques applied to different biomasses from agriculture forestry and waste, subjected to different prerequisites to obtain samples with different particle size treatments. In this study

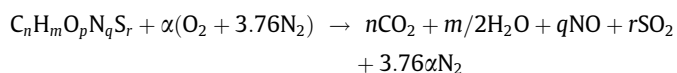
samples are classified by a graph of thermal sensitivity, differentiating the degree of self-ignition risk. The influence of the physical preparation of the samples and the importance of the chemical composition is analysed, noting that parameters from ultimate analysis are highly correlated with thermal susceptibility.

2. Materials

The experiments have been conducted with nineteen samples obtained from eleven biomass products of agriculture, forestry and waste residues. All of the samples are commercial biomasses suitable for gasification processes. These biomasses were exposed to different ground processes obtaining the final studied products [23]. By using different ground processes is intended to obtain dust particles of various sizes to study the self-heating process first and the self-ignition process after.

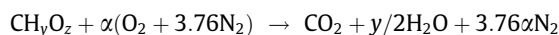
Particle size of biomass should be considered since it affects the behaviour and the kinetics of several processes, such as gasification [24]. It should also be noted that the variety of particle shapes results in different surfaces, an essential parameter in the process of heating and heat transfer [25,26]. Therefore, granulometry is expected to play a certain role in the self-heating process, thus conditioning all the parameters measured. Consequently, the particle size distributions were measured using a Malvern Mastersizer laser diffraction apparatus. Table 1 presents the type of grounding used and the particle size of the nineteen analysed biomasses, expressed as mass median diameter $d[0.5]$ and the size of particle below which 10% and 90% of the sample lies, $d[0.1]$ and $d[0.9]$, respectively. Table 2 shows the proximate and ultimate analysis of samples, obtained by conventional analytical techniques.

From the chemical composition is possible to obtain the approximate formulas of solid fuels and state their stoichiometric combustion reactions:



where α is determined by $\alpha = n + m/4 - p/2 + q/2 + r$.

A simplified formula for biomass can be estimated neglecting N and S contents, establishing an empirical formula CH_yO_z .



where α is now determined by $\alpha = 1 + y/4 - z/2$ and the number of air moles is $\alpha/0.21$.

Coefficients y and z represent, respectively, the atomic ratios H/C and O/C, which can be easily determined from the ultimate analysis, thus establishing the empirical formula of the biomass. From the balanced combustion equation in air, the stoichiometric air to fuel mass ratio, A/F , can be obtained for a typical molecular mass of air of 28.84 g per mole:

$$[A/F]_{\text{stoic}} = (\alpha/0.21 \cdot 28.84)/(12 + y + 16z)$$

Also, for an air density of 1200 g/m³, dust concentrations for stoichiometric concentrations (equivalence ratio $\Phi = 1$) can be obtained [27]. These are in the range 177–216 g/m³, two or three times below typical concentrations producing the maximum explosion effects [28]. Table 3 shows these stoichiometry parameters for the nineteen biomass samples.

According to the obtained values for H/C and O/C, the experimental formulas for these products would range from $CH_{1.29}O_{0.52}$ for Grape Seed Meal to $CH_{1.49}O_{0.72}$ for Brassica.

Agroforestry biomass are generally heterogeneous products structurally made of cellulose, hemicelluloses, lignin and other components at low concentrations such as proteins, acids, salts and minerals [29]. The set of cellulose and hemicelluloses is known as holocellulose and represents two-thirds of the dry matter. It is

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