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Oxidative Pyrolysis of Agricultural Residues in Gasification and Carbonization Processes

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Abstract. Oxidative pyrolysis, in which heat is provided by the partial oxidation, occurs in many fixed bed reactors of air staged gasification and advanced carbonization processes. In these reactors, an Oxidation Zone (OZ) propagates and separates the virgin biomass from the produced char, and self-sustains the process. To investigate the energy transfer efficiency and to characterize the OZ features, oxidative pyrolysis was performed in a 20 cm diameter fixed bed reactor with Wheat straw pellets. Temperatures and biomass bed height were measured continuously during the experiment. The OZ propagated at 0.41 cm/min and consumed about 11 % of the biomass. The role of bed compaction on the effective propagation velocity was highlighted. Char yield was measured to 28.6 % and char accounted for 39.56 % of the total energy content of the Wheat straw. Maximum temperature in the bed reached 780 °C. This paper proposes a new insight into the OZ features when it propagates in a biomass fixed bed.

1. Introduction

Agricultural residues must be disposed in order to avoid contamination, pest growth, occupying large expanses of land and hindering agricultural work. In addition, agricultural residues can be exploited for recovering specific components or useful energy. Therefore, valorization of agricultural residues for energy purposes has multiple advantages: minimizing the environmental impacts, increasing economic benefits for the farmers, and reducing the reliance on fossil fuels [1], [2].

Holding a huge potential and the carbon-rich composition, agricultural residues are being considered as the major contributors of resources for energy and material productions. The global annual production of agricultural residues of the six most important crops (barley, maize, rice, soybean, sugar cane and wheat) in 227 countries and territories of the world is reported to be $3.7x10^{12}$ kg of dried matter. This residues production is proportional to crop yields [1], [3].

Thermo-chemical conversion, i.e. combustion, gasification and pyrolysis can convert agricultural residues into variety of useful energy types such as heat, electricity and materials like charcoal, syngas and bio-oil. Pyrolysis for charcoal production, carbonization, is one of the most widely used technologies. It is normally known as the thermal decomposition of organic components of biomass in an inert gas atmosphere, at moderate temperature. In such technology, the external energy source is needed to provide heat for the pyrolysis reactions. However, some current studies have shown that the partial oxidation of biomass and products, so called *oxidative pyrolysis*, is an effective way to provide

energy to the process [4], [5]. Oxidative pyrolysis allows the autothermal operation; and contributes to reduce complexity of the system and operating cost.
Here, we characterized the oxidation features in terms of propagation velocity, temperature, char yield, equivalence ratio and energy biomass conversion to pyrolysis products.

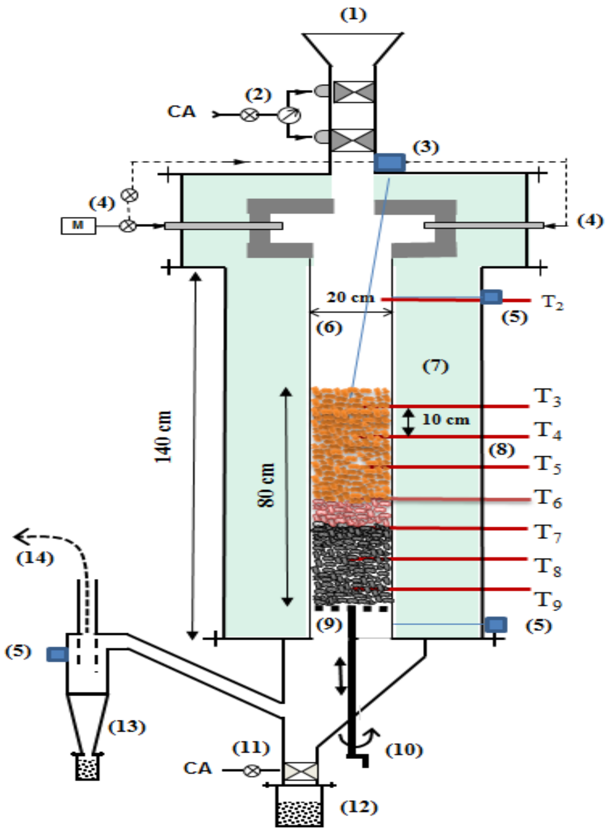
2. Material and equipment

2.1. Material
In this study, we used Wheat straw as a specific agricultural residue (Table 1). The pellet form was used to avoid the bridging problem occurring in stratified fixed bed reactors [6]–[8] and to ensure the homogeneity in the biomass bed properties.

Table 1. Wheat straw pellets properties

Density (kg/m^3)	Moisture (wt%)	Proximate analysis (wt%, d.b.)			Ultimate analysis (wt%, d.a.f.)				LHV, (MJ/kg)
		Volatile mater	Ash	Fixed carbon	C	H	N	O	
549 ±1.42	8.75 ±0.05	78.75 ±1.89	8.17 ±0.09	13.08 ±1.89	58.91 ±0.18	5.07 ±0.04	1.10 ±0.10	34.92 ±0.24	17.45 ±0.12

2.2. Equipment description
Oxidative pyrolysis was studied in a fixed bed reactor (Figure 1). It consists of a 310 type refractory steel tube (20cm I.D.; 160 cm L) surrounded by a 30 cm thick refractory wool insulation. Thermocouples are installed every 10 cm inside the bed, allowing continuous measurement of temperature. In addition, a laser beam device is placed above the biomass bed to continuously measure the bed height during experiments allowing the calculation of the bed compaction.



1- Hopper
2- Pneumatic valves
3- Laser beam device
4- Air entrances
5- Transducers
6- Reactor tube
7- Insulation
8- Thermocouples
9- Grate
10- Char extraction system
11- Gate valve
12- Char tank
13- Cyclone
14- Post-combustion system
M- Flowmeter
CA- Compressed Air

Figure 1. The fixed bed reactor

The experimental procedure consists in igniting the OZ at the bottom of a 70 cm high biomass bed and following its upward propagation (about 10 cm of charcoal was used for the ignition). For this purpose, air flow is supplied at the top of the reactor and passes down through the biomass bed. As an example, evolution of temperatures at the different thermocouples presented in figure 2a shows the propagation of the OZ through a Wheat straw pellets bed while figure 2b shows the bed height reduction during the test.

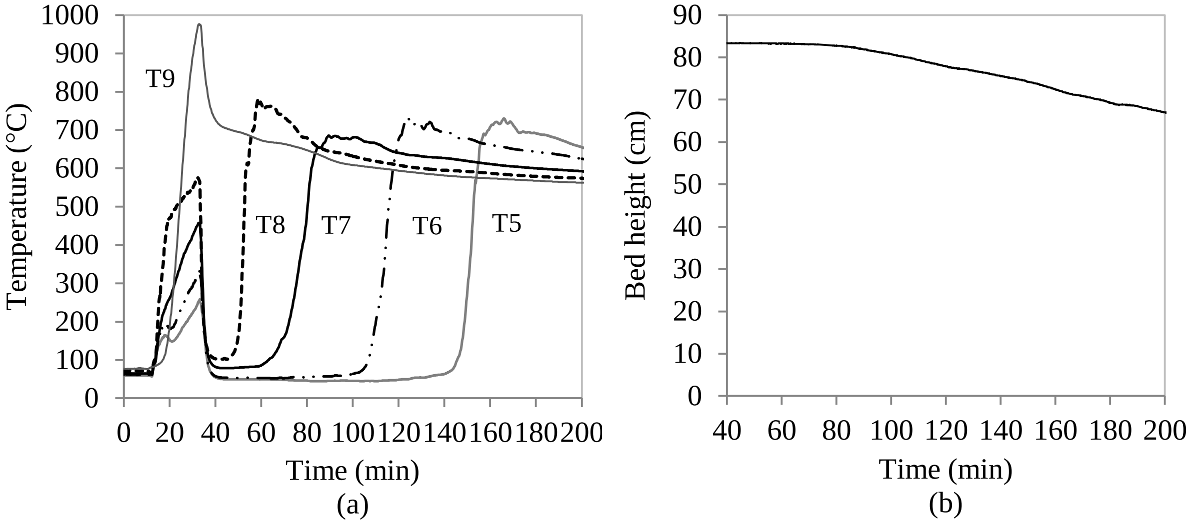


Figure 2. Typical experimental results with an air flux of 0.022 kg/m^{2}s:

(a) Temperature profile and (b) Bed height reduction

In Figure 2a, first the temperature in T9 reach 950-1000 ℃ after about 30 mins following the ignition. Then, the air flux was reduced to the studied value of 0.022 kg/m^{2}s involving a decrease of the temperature tends to about 600 ℃ up to the end of the experiment. Then, the oxidation zone propagates upward from T9 position to the top of the bed, in T5 position. The passage is observed by the increase of each temperature measured by thermocouples located all along the biomass/char bed each 10 cm. Thus, the temperature profiles versus time can be used for determination of the OZ propagation velocity in the biomass bed.

The bed height remains constant up to 80 mins, then, continuously reduces up to the end of the test (Figure 2b). This fact reveals that the oxidation zone propagates in a stationary regime only after reaching T7. Thus, in further discussion, we focus on the zone behavior from T7 to T5, i.e. 25cm to 45 cm respectively from the bottom of the bed. The bed height measurement allows the determination of the bed compaction velocity, and thus the biomass consumption rate.

3. Method for characterizing the oxidation zone features

From these temperature and bed height measurements, a new method has been proposed to calculate the upward propagation velocity of the oxidation zone. In this reverse smoldering configuration, the OZ propagates upward while the bed compaction makes biomass going down into the oxidation zone. The bed compaction velocity needs to be taken into account when determining the biomass consumption rate (Figure 3).

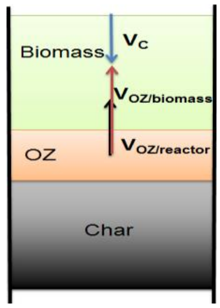


Figure 3. Simplified sketch illustrating OZ velocities and compaction velocity in fixed bed

We define $V_{OZ/reactor}$ (cm/min) as the apparent upward velocity. It is the OZ velocity related to the reactor, the one we observe from outside of the reactor. It is measured by the temperature evolution inside the reactor as described in next section. $V_{OZ/biomass}$ (cm/min) is defined as the velocity of the oxidation zone related to biomass, which is an effective velocity of propagation of the oxidation zone in the biomass. V_C (cm/min) is defined as the compaction velocity; it is measured by the laser during experiments and allows the calculation of Voz/biomass by following equation:

$$V_{OZ/biomass} = V_{OZ/reactor} + V_C \quad (1)$$

Here, we assume that the bed compaction only occurs within the oxidation zone. In other words, there is no compaction in the bed of biomass and in the bed of produced char.

3.1. Measurement of $V_{OZ/reactor}$

The position of the oxidation zone at the elevation of a given thermocouple Ti is indicated when dTi/dt is maximum. We define t^i_{OZ} as the time coresponding to the passage of the OZ at that elevation (Figure 4).

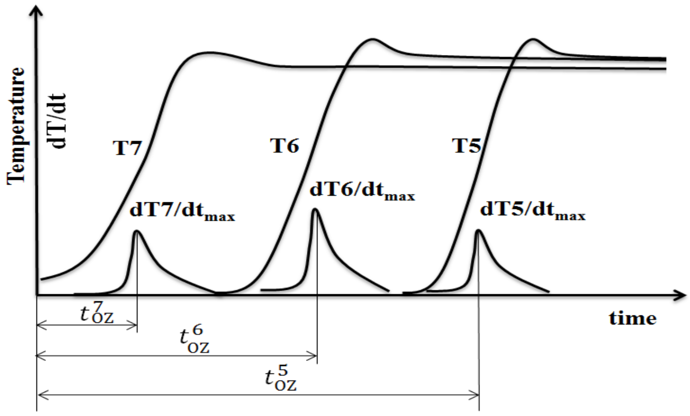


Figure 4. OZ passage time determination.

The position of the oxidation zone in the reactor versus time can be plotted (Figure 5). The upward propagation velocity of the OZ can be determined from the derivative of the position function. For this purpose, a fitting of the experimental data was performed (Figure 6).

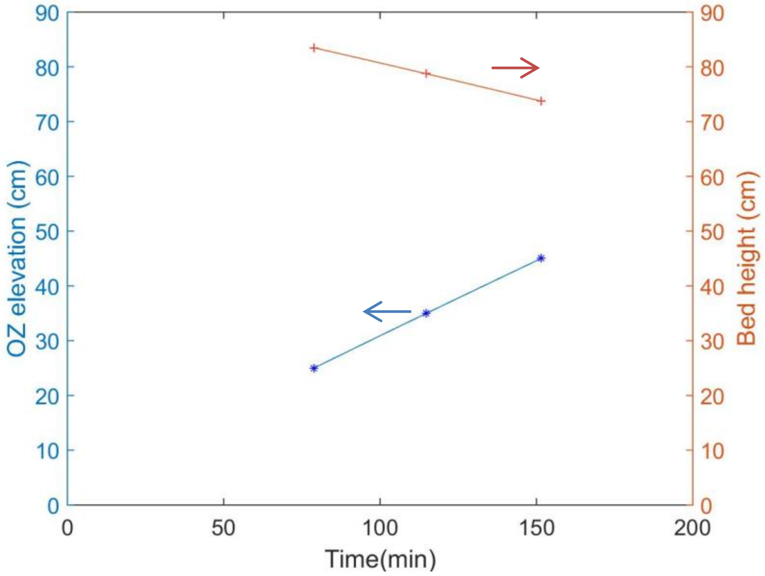


Figure 5. The evolution of the OZ propagation rate along a Wheat straw bed (air flux: 0.022 kg/m^{2}s).

As shown in Figure 5, the oxidation zone reached T7 position (Z=25cm) at around 80 mins, then propagated upward and reached T6 (Z=35cm) at 118 mins and T5 (Z=45cm) at 156 mins. The total bed height reduced from 82 cm when OZ arrived at T7 (25 cm) to around 74 cm when OZ arrived at T5 (45 cm).

3.2. Measurement of the biomass consumption and char production

The hypothesis that the compaction happens only within the oxidation zone allows the estimation of the apparent OZ char velocity ($V_{oz/char}$):

$$V_{OZ/char} = V_{OZ/reactor} \tag{2}$$

The biomass consumption rate then is calculated as following equation:

$$\dot{m}_{biomass} = V_{OZ/biomass} * A_{reactor} * \rho_{biomass} \tag{3}$$

Char production rate:

$$\dot{m}_{char} = V_{OZ/reactor} * A_{reactor} * \rho_{char} \tag{4}$$

ρ_{char} is the density of char which was measured about 236 kg/m^3.

3.3. Equivalence ratio (ER)

The ratio of air to biomass is written:

$$Air: biomass = \frac{Air\ mass\ flux}{Biomass\ consumption\ rate} \tag{5}$$

Thus, the ER is classically defined as follow:

$$ER = \frac{Air: biomass}{Air: biomass_{stoichiometry}} \tag{6}$$

Note that the Air: biomass stoichiometry is 5.38 calculated from the ultimate analysis (Table 1) of Wheat straw pellets.

4. Results and discussions

4.1. Propagation velocities of the oxidation zone

Propagation velocities include the apparent velocity ($V_{OZ/reactor}$) and the effective velocity ($V_{OZ/biomass}$). In figure 6, the apparent velocity of the oxidation zone was shown about 0.28 cm/min while the bed compaction velocity was found to be 0.13 cm/min. As a consequence, the effective velocity of the oxidation zone in the biomass bed was calculated to 0.41 cm/min. Bed compaction and propagation velocities were almost constant during the test period, which confirm that a stationary regime was reached in the zone from 25 cm to 45 cm in the reactor (from T7 and T5).

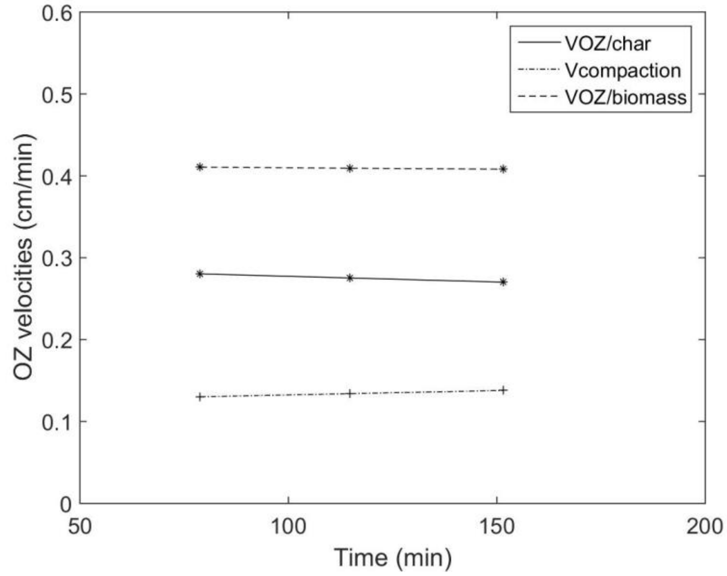


Figure 6. OZ propagation velocities with an air flux of 0.022 kg/m^{2}s.

These results show that in downdraft fixed bed configuration, the bed compaction accelerated the propagation of the oxidation zone. However, few studies [9], [10] have taken the bed compaction into account when calculating the propagation velocity of the oxidation zone.

4.2. Temperature

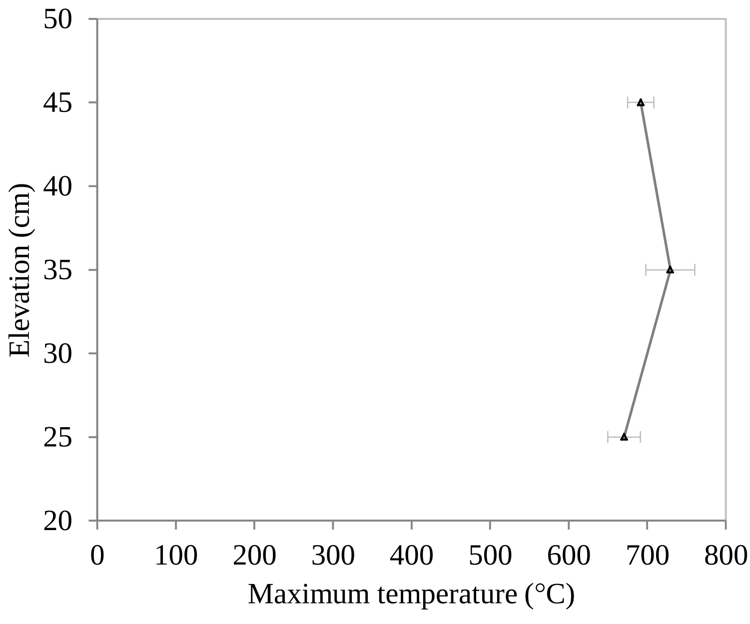


Figure 7. Maximum temperature profile with an air flux of 0.022 kg/m^{2}s.

The maximum temperature (Figure 7) in the bed of Wheat straw pellets oscillated between 650 ℃ and 780 ℃ and no hot spot temperature along the bed was observed. Temperature is a major influencing factor regarding pyrolysis products, yields and composition. It is reported that pyrolysis temperature is increased, the char yield decreased [11], [12].

4.3. Biomass consumption rate, char production rate and equivalence ratio

Propagation velocities allow the calculation of the biomass consumption rate and the char production rate (Figure 8). In our operating conditions, biomass consumption rate was calculated to 4.2 kg/h and the char production rate to 1.2 kg/h. The char yield defined as the ratio of char production rate over biomass consumption rate was 28.6 %. This result shows that char yield in the oxidative pyrolysis was in line with the typical slow pyrolysis processes [13].

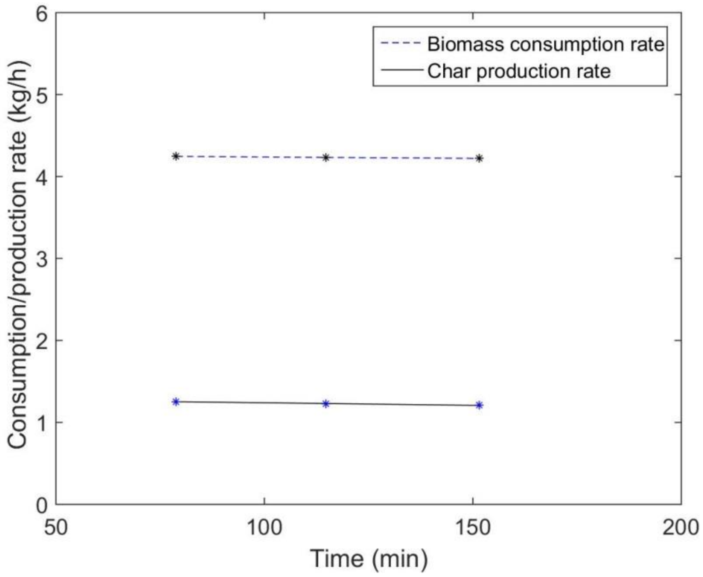


Figure 8. Biomass consumption rate and char production rate with an air flux of 0.022 kg/m^{2}s.

The energy transferred from biomass to the char was calculated from the measurements of char yield and the lower heating value (LHV) of charcoal and biomass.
The LHV was measured to 24.14 MJ/kg and 17.45 MJ/kg for charcoal and biomass, respectively.

$$E_E = \frac{\dot{m}_{char} * LHV_{char}}{\dot{m}_{biomass} * LHV_{biomass}} = 39.56\ \% \qquad (7)$$

Energy transfer to the char was about 39.56 %. The rest of energy in biomass was transferred to the other products such as permanent gases and liquid fuels.
Finally, the ER of our autothermal pyrolysis process was calculated to be 0.11. It means that the oxidation zone consumed 11 % biomass to provide heat for self-sustain the process.

5. Conclusions

The aim of this work was to investigate the oxidative autothermal pyrolysis in fixed beds of agricultural residues pellets. The OZ features in terms of propagation velocity, temperature, char yield and equivalence ratio were discussed. Regarding the air mass flux of 0.022 kg/m^{2}s, the oxidation zone propagates at an effective velocity of 0.41 cm/min in the biomass bed. The maximum temperature oscillated between 650°C and 780°C. The OZ consumes 11 % of the Wheat straw to provide energy for the autothermal process. Char yield and energy transferred from biomass to the char were measured to 28.6 % and 39.56 %, respectively.
These results suggest the direction for the valorisation agricultural residues for energy purpose by oxidative pyrolysis. The future work will focus on the characterization the oxidation zone features in fixed bed of various biomass types.

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