CHEMICAL ENGINEERING TRANSACTIONS
VOL. 65, 2018
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Guest Editors: Eliseo Ranzi, Mario Costa

ISBN 978-88-95608- 62-4; **ISSN** 2283-9216
DOI: 10.3303/CET1865060

Biocarbon Pellet Production: Optimization of Pelletizing Process

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Biocarbon is charcoal produced as a by-product of fast pyrolysis and gasification or as the main product of slow pyrolysis. The biocarbon can be used as a fuel in combustion systems both at large scale and small scale. The University of Perugia is collaborating with SINTEF IN Norway regarding biocarbon combustion tests in pellet boilers, adapting the fuel and the air mass flows to the new fuel. Overall, the two critical aspects are: 1) the production of pellet from biocarbon and 2) its combustion performance in small boilers. The objective of the work is to optimize biocarbon pellet production. To produce this new kind of pellet the moisture content and the use of additives have to be carefully adjusted and optimized. A Box-Behnken experimental scheme has been used for this purpose. It was implemented in the software Minitab 17. Fifteen pelletization tests have been performed with mixtures of biocarbon, sawdust and water in different proportions. The durability of the obtained pellet was measured. The optimal mixture was found to be: 30%w/w moisture, 30%w/w sawdust and 40%w/w biochar. The obtained pellet had both higher durability and heating value (23,057 kJ/kg).

1. Introduction

Pyrolysis of biomass produces three main products (Corbetta et al. 2014 and Gentile et al. 2017): solid (charcoal or biocarbon), liquid (biooil) and gaseous (pyrogas). Each of the three products has interesting energy content and can be used as a fuel and for the production of biochemicals (He et al. 2018 and He et al. 2017 and Wang et al. 2017). The pelletization of charcoal, obtained from biomass pyrolysis, is interesting to improve the following characteristics of the fuel: bulk density, energy density, transportation costs, storage costs, co-firing performances with coal; as reported in Hu et al. (2015). Different materials have been tested in the pelletizing process: Peng et al. (2015) worked with torrefied wood, Hu et al. (2015) and Hu et al. (2016) used pyrolysis char; Reza et al. (2014) and Reza et al. (2012) worked with hydrochar. There is a general agreement on the fact that sawdust has a good performance as an additive for biochar pelletization. According to Peng et al. (2015) also moisture and the size of the material can play an important role. Peng et al. (2015) performed pelletizing tests with torrefied biomass, using different binders - such as sawdust, starch and lignin. The pelletizing conditions were standardized: die temperature equal to 110°C, compression pressure equal to 125 MPa, holding time equal to 1 minute and maximum pressure equal to 125 MPa. A press machine (Measurement Technology Inc., USA; Model: MTI 50 K) was used to compress ground biochar into a single pellet. It was found that the addition of sawdust (especially in the ratio of 30%w/w) decreases the energy demand for the pelletization process and increases the hardness of the obtained pellet. Hence the addition of sawdust in the pelletization tests has a very positive impact.

The key process during pelletization is the softening of the lignin (contained in the sawdust) during compression and parallel heating, as this grants a high elastic modulus and good bonding properties of pellets (see Hu et al. 2015). Hu et al. 2015 arrived at the conclusion that moisture is a fundamental parameter in biochar pelletization. An optimal value of 20%w/w of moisture is reported together with a value of 10%w/w binder. The best binder was found to be lignin. So it can be inferred that the moisture of the material is maybe

Please cite this article as: Bartocci P., Barbanera M., Skreiberg O., Wang L., Bidini G., Fantozzi F., 2018, Biocarbon pellet production: optimization of pelletizing process, Chemical Engineering Transactions, 65, 355-360 DOI: 10.3303/CET1865060

less important for torrefied biomass but it is fundamental for biochar obtained from pyrolysis. In another study Hu et al. 2016 analyze the influence on the pelletization process of the pyrolysis temperature. Through an accurate Scanning Electrone Microscope analysis Hu et al. 2016 and Hu et al. 2015 analyzed the mechanism that regulates biochar pellet densification quality. It was found that with pyrolysis temperatures equal to 650°C the optimal quantity of water is about 35%w/w, while the binder (mainly lignin) can be about 10%w/w. Dealing with the main processes occurring during biochar pelletization, it must be considered that water and lignin can promote pelletization of carbonized brittle and fine biochar particles derived from pyrolysis at 650°C, acting as binders. Lignin which is a randomly cross-linked polymer of phenylpropane units, has a glass transition behaviour in the temperature range of 137-157°C (see Reza et al. 2012). The presence of water during the compacting process decreases the glass transition temperature of lignin, so with 35%w/w of water in the raw material and the pressure of 128 MPa, lignin forms solid bridges between the biochar particles, which enhance the compressive strength of the biochar pellet.
While several laboratory tests have analysed the pelletising process using single channel press machines, continuous production of biocarbon pellet has never been analysed. To fill this gap, the authors have focused their attention on a continuous pelletizing machine, with mass flow of 40 kg/h. To achieve satisfactory durability of the biocarbon pellet was proven to be quite hard. It is almost impossible to produce it without additives (for example sawdust) and a proper optimization of the moisture content. The aim of the work has been to identify the optimal quantity of water and additives in a continuous biocarbon pelletizing process. This was achieved using a Response Surface Methodology (RSM) to identify which are the pelletizing parameters that provide the highest durability.

2. Materials and methods

The material has been taken from the IPRP pilot plant (Integrated Pyrolysis Regenerated Plant) designed and realized at the university of Perugia and described in previous works (Bartocci et al. 2016). Pelletizing tests have been performed at the Laboratories of the Biomass Research Centre, University of Perugia using biochar produced from local biomasses. Design of Experiment was implemented in the software Minitab 17. Fifteen pelletization tests have been performed with mixtures of biocarbon, sawdust and water in different proportions. The pelletization tests have been based on a Box-Behnken scheme with three factors (saw dust ratio, moisture ratio and biochar size), three levels and three central points. The parameter to optimize was the final durability of the obtained pellet. The pelletizing machine used in the tests is produced by Smartec and the model is PLT 100. The main features are the following: 40 kg/h product mass flow, 6 mm pellet diameter, 4 kWe engine power, 100 kg machine weight. In this work about 10 kg of biochar was taken from the IPRP plant in Terni, Italy. Sawdust was used as an additive (see Figure 1).

Figure 1: Raw materials used in the pelletization tests: biochar (left) and sawdust (right)

The standards used for the characterization of the raw materials are reported in Table 1. The used charcoal was produced through a slow pyrolysis process at a final temperature of 600°C. The raw material was woodchips produced from Umbrian coppice woods.
The size of the biochar was quite big so it was milled with a Retsch SM 2000 cutting mill, using different sieves (2, 6 and 10 millimeters). The sawdust was bought from local retailers. The raw materials and the obtained pellet were characterized performing proximate analysis, ultimate analysis and calorimetry. The characterization of the two starting materials was performed at the laboratories of the Biomass Research Centre. Results are shown in Table 2 (d.b. stands for dry basis).

Table 1: Standards used for product characterization

Analytical procedure	Standard	Instrument
Quartering	BS5309-1:1976	Manually
Determination of moisture content	UNI EN14774-2:2010	LECO TGA 701
Determination of ash content	UNI EN14775:2010	LECO TGA 701
Determination of volatile matter	UNI EN15148:2009	LECO TGA 701
Determination of carbon, hydrogen, nitrogen	UNI EN15104:2011	LECO Truspec CHN
Determination of calorific value	UNI EN14918:2009	LECO AC-350
Determination of pellet durability	UNI 15210-1:2010	Holmen Tester TekPro

Table 2: Raw materials characterization

Characteristics	Sawdust	Charcoal
Moisture (%w/w)	8.95	4.50
Volatiles (%w/w d.b.)	81.66	22.09
Ashes (%w/w d.b.)	0.65	7.86
Fixed carbon (%w/w d.b.)	17.69	70.05
C (%w/w d.b.)	46.40	73.00
H (%w/w d.b.)	6.62	3.26
N (%w/w d.b.)	0.87	1.27
O (%w/w d.b.)	46.11	22.47
Higher Heating Value - HHV - (kJ/kg)	19,806	28,532

3. Results and discussion

3.1 Results of the pelletizing experiments

In Table 3 the final durability of the obtained pellet is reported, for each pelletizing test contained inside the Box-Behnken scheme. The response of the experimental design was not always satisfactory.

Table 3: Raw materials characterization

Test number	Sawdust (%w/w)	Particle size (mm)	Moisture (%w/w)	Durability (%)
1	0	2	20	n.a
2	0	6	10	n.a.
3	0	6	30	85
4	0	10	20	n.a.
5	15	2	10	n.a.
6	15	2	30	90
7	15	6	20	n.a.
8	15	6	20	n.a.
9	15	6	20	n.a.
10	15	10	10	n.a.
11	15	10	30	92
12	30	2	20	n.a.
13	30	6	10	n.a.
14	30	6	30	95
15	30	10	20	n.a.

The images of the used mixtures are shown in Figure 2. Each sample was represented by a mixture of about 400 g weight. From Table 3 it can be seen that no pellet could be produced with mixtures containing a moisture which was lower than 30%w/w (the particles did not aggregate and exited the machine in powder form). Only mixtures with initial moisture of 30%w/w gave durable pellet, while the other mixtures passed through the pelletizing machine producing a sort of powder. When the moisture of the mixture was enough to produce the pellet the yield of pellet, given by the ratio between the pellet produced and the 400 grams used as input material was about 40%w/w. This means that about 60%w/w in weight of the used mixture remained inside the pelletizing machine.

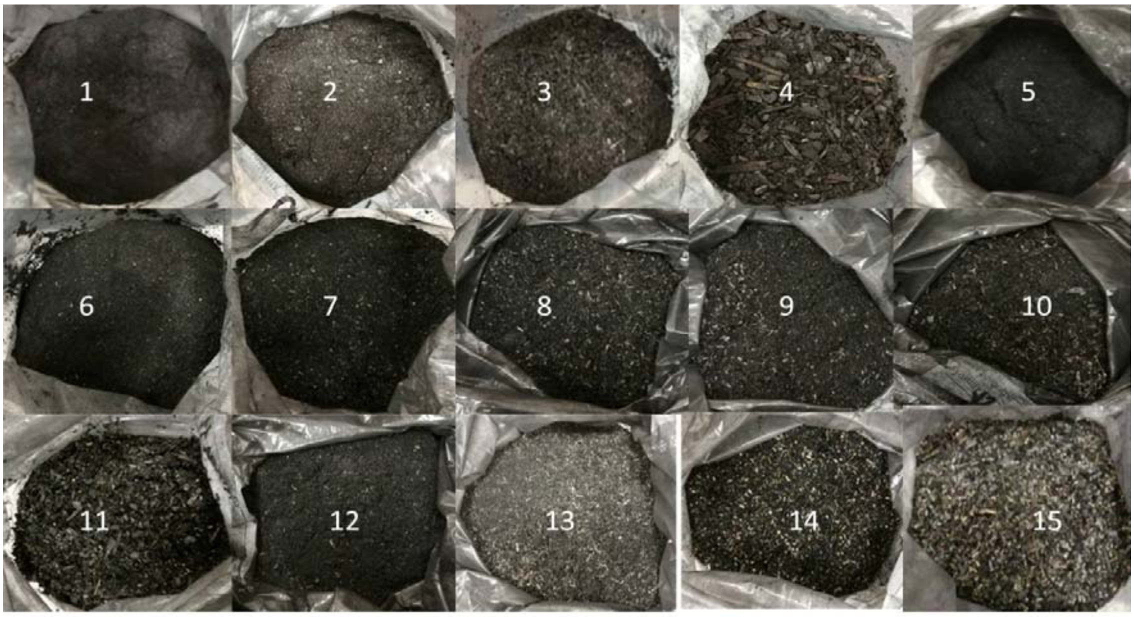

Figure 2: Mixtures used in the pelletization tests

The explanation for the results shown in Table 3 is that on one hand the biocarbon needed water to aggregate and form pellet with an acceptable mechanical strength; on the other hand too high moisture content in the pellet resulted in bad cohesion. Hence there is an optimal moisture content to grant good durability. The content of water in the raw material influences also the temperature of the die. If the temperature of the die is too high the compression force needed to produce the pellet decreases and so decreases also the durability of the pellet. It was found that during the tests it is highly recommended to start with high moisture content of the mixture (to avoid the heating of the die) and to recycle the material in the output for 5 to 10 times to decrease its moisture gradually and obtain good mechanical strength of the material.

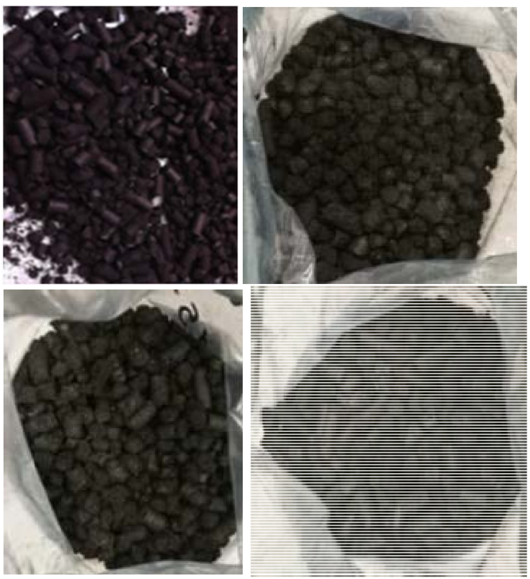

Figure 3: Obtained pellet from test 3 (up-left), test 6 (up-right), test 11 (down-left) and test 14 (down-right)

This is a commonly used procedure in literature, with small pelletizing machines. It has to be considered in fact that Young's compression modulus for charcoal (which ranges from 4 to 9 GPa, according to Emmerich and Luengo 1996) is higher than that of wood (which ranges between 0.5 and 1 GPa, according to Holm et al. 2006). Another aspect to take into consideration is that the friction coefficient of the material increases with the increase in moisture content. The images of the obtained pellet are presented in Figure 3.

3.2 Biocarbon pellet characterization

The chemico-physical characterization of the obtained pellet is shown in Table 4. It can be seen that pellet durability is proportional to the pellet moisture, which is influenced by both: the temperature of the die and the porosity of the material contained in the mixture. In particular biochar is able to retain more water than wood, due to its higher porosity. Together with the final moisture of the mixture, another parameter which influences the durability of the pellet is the size of the biochar particle (usually the optimal size for the pellet made with sawdust was found to be about 4 mm). As it can be seen from Table 4, the pellet obtained from mixture 14 (that is the mixture obtained with 30%w/w water, 40%w/w biochar and 30%w/w sawdust) has the highest calorific value (equal to 23,057 kJ/kg) and also the lowest ash content (equal to 4.77 %w/w). The low ash content can make this fuel interesting also for household heating in conventional pellet boilers and stoves. This fuel is interesting also for cofiring with coal. The Higher Heating Value of the obtained fuel is about 81%w/w that of the initial biochar. The heating value (wet basis) decreases due to the presence of sawdust as a binder, but it remains quite high, due to the lower moisture content of the final product.

Table 4: Raw materials characterization

Characteristics	Mixture 3	Mixture 6	Mixture 11	Mixture 14
Moisture (%w/w)	23.10	15.06	14.05	7.11
Volatiles (%w/w d.b.)	22.09	34.86	34.86	47.62
Ashes (%w/w d.b.)	7.86	6.32	6.32	4.77
Fixed carbon (%w/w d.b.)	70.05	58.83	58.83	47.61
C (%w/w d.b.)	56.21	57.21	57.88	57.29
H (%w/w d.b.)	5.07	5.05	4.98	5.15
N (%w/w d.b.)	0.98	1.01	1.02	1.02
O (%w/w d.b.)	37.75	36.74	36.13	36.54
HHVw.b. (kJ/kg)	21,970	22,663	22,929	23,057

Figure 4 shows a regression analysis in which it is demonstrated that there is a correlation between the moisture of the pellet and its durability. The coefficient of determination of the linear regression (R^2) is about 0.96. These are obviously preliminary data which have been confirmed by the following experimental schemes. Further check of the presented results should be performed with a mechanistic model of the pelletizing process, such that proposed by Aalborg university and Andritz Feed and Biofuels (Nielsen 2016).

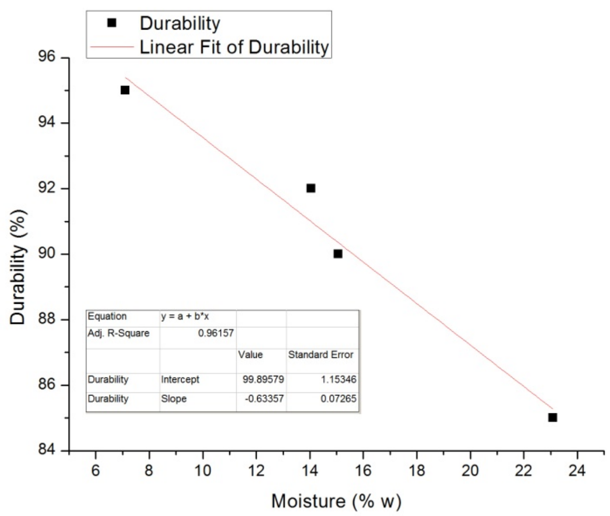

Figure 4: Correlation between pellet moisture and durability

4. Conclusions

This paper presents the results of pelletization tests on biochar produced from slow pyrolysis at the final temperature of 600°C. A Box-Behnken experimental scheme was executed based on 15 tests with three factors (saw dust ratio, moisture ratio and biochar size), three levels and three central points. All the samples with moisture less than 30%w/w in weight did not produce pellet but only dust and some aggregates. The mixtures number 3, 6, 11 and 14, with 30%w/w moisture, produced durable pellet. The moisture content of the final product was inversely proportional to the sawdust content of the mixture. This was due to the different porosity of sawdust and biochar. The durability of the pellet was inversely proportional to the moisture content of the final product. The pellet obtained from mixture 14 (with 30%w/w water, 40%w/w biochar and 30%w/w sawdust) had the highest calorific value (equal to 23,057 kJ/kg) and also the lowest ash content (equal to 4.77 %w/w).

Acknowledgments

The authors would like to acknowledge eng. Michele Oligarchi, eng. Mirco Cesca and eng. Silvia Falcinelli for the help in pelletizing tests.

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