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Additional Information

Influence of raw material composition of Mediterranean pinewood on pellet quality

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Abstract

The lack of management and economic value of forest ecosystems in Mediterranean areas, mainly dominated by pines (*Pinus halepensis* Mill. and *Pinus pinaster* Ait.), together with a continuous growing of forests on marginal agricultural land have led to an increased forest fire risk and devaluation of natural resources in less favoured rural regions. The use of residual forest-based biomass as value added biofuels like pellets can reverse this situation. Nevertheless, there is an industrial need of knowledge of the influence of raw material composition on pellet manufacturing quality for the main Mediterranean softwood species. In this research influencing pellet quality variables such as contents of moisture, ash, fine particles, chlorine and sulphate, but also mechanical durability, bulk density and net calorific value have been analysed following EN standards. The obtained results demonstrate that it is possible to obtain high quality pellets from barked and debarked logs as well as from branches from *Pinus halepensis* and *Pinus pinaster*. This implies that the abundant forest biomass residues in the western Mediterranean region of Europe can be transformed into high added value solid biofuels, opening new opportunities for local industries.

Keywords: Mediterranean pine; wood pellets; bark content; biofuel quality; residual biomass

1. Introduction

Pinus halepensis Mill. and *Pinus pinaster* Ait. occupy big extensions along the Mediterranean basin and represent the only wood source and forest canopy in many Mediterranean countries (Prada 2008). In Spain, both pines are the more abundant softwood species with 1.770.000 ha and 1.060.000 ha respectively (IFN3 2008), as consequence of natural expansion and big afforestation accomplished during the second half of the 20th century. Despite forest fires and other risks as plagues, due to the generalised abandonment of rural areas and depopulation, e.g. in the Valencian region the forest areas are continuously growing around 3.000 ha annually (PATFOR 2012). Consequence of all these factors together with the lack of forest management (due to the low economic revenues generated by these type of forests of low wood quality and difficult orography) is that biomass is being yearly accumulated (Marraco 1991) without an economic use and forest fires have increased both in number and in severity (Guardia 2000).

The use of forest-based biomass as bioenergy represents an opportunity to mobilize a big potential of forest resources and residues that have no market value (AEBIOM 2015). Moreover, this offers a key opportunity for the Mediterranean forest sector and the rural societies to manage their own resources, but also to reduce its dependence on non-renewable energy sources, allowing a reduction of forest fire risks and improving forest ecosystem resilience (PROFORBIOMED 2014).

In that sense, pellets are nowadays an important and competitive renewable biofuel, a compact and standard product with a high calorific value per weight unit due to its low humidity and its high density (Bhattachaya et al. 1989), that allows a reduction of four times the volume of biofuel compared to woodchips, enabling easier logistic processes and lower transport costs (Fernández 2009). In Spain, according to AVEBIOM (2014) pellet production capacity installed in 2015 was of 1.34 million tonnes a year produced by a total of 60 pellet plants, with an expected production of 475.000 tonnes. Therefore, there is an increasing need for raw material supply.

Traditionally, the raw material for wood pellet production are softwood woodchips, because they have lower density and therefore are easier than hardwoods to process (Filbakk et al. 2011). Moreover, stem wood is preferred to other tree fractions (bark, roots, crown and branches) because required material homogeneity and predictability in combustion. Thus, generally stems are debarked before being processed in sawmills or chipped. But even though preferred recovered raw materials for production of wood pellets, such as cutter shavings and sawdust, are used to the maximum extent (Stahl and Berghel 2011), the increase of pellet demand entailed the supply of pure wood to be insufficient and other raw materials have been used. In order to widen the raw material base and reduce pellet production costs, it would be advantageous to utilize the whole log, including branches and bark (Toscano et al. 2013).

Despite there exist numerous scientific studies about pellet quality and influence of raw material, especially on Northern European softwood species with an extended industrial use such as Scots pine *Pinus sylvestris* and Norway spruce *Picea abies* (Arshadi et al. 2008, Samuelson et al. 2009, Filbakk et al. 2011, Lestander et al. 2012), there is a lack of knowledge of studies related with the most important Mediterranean softwood species such as *Pinus halepensis* and *Pinus pinaster* (Lerma-Arce 2015).

A deeper scientific and technical knowledge on pellet manufacturing based on full-tree chipped material of these species would help to widen the raw material supply and to reduce costs in pellet manufacturing processes. Moreover, it would contribute to mobilize non-used forest resources and to reduce drastically the risk of large forest fires by

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reducing biofuel accumulation, particularly in a scenario of very hot summers derived of the process of climate change in the Mediterranean basin (FAO 2011).

2. Objectives

The general aim of this research has been to analyse the influence of the raw material composition on the quality of pellets obtained from different woodchips assortments of two representative Mediterranean pinewood species, *Pinus halepensis* and *Pinus pinaster*.

To achieve this aim, the specific objectives of the research are:

- a) to manufacture pellets from different woodchips assortments: debarked stem logs, non-debarked stem logs and branches,
- b) to analyse pellet quality and the relationship between quality factors and their possible interdependence,
- c) to study the maximum tolerated bark content for each pellet assortment to accomplish with the maximum tolerated ash content according to European quality standards.

3. Materials and methods

3.1 Materials

The material was taken from a 63 years old afforested stand of *Pinus halepensis* and *Pinus pinaster* in the forest district V074 "Navalón" of Enguera (Valencia, Spain). As shown in figures 1a and 1b, this forest is very representative for the most important forest type in the Mediterranean area of the Iberian Peninsula (IFN3 2008, Lerma-Arce 2015).

...Figures 1a and 1b about here...

3.2 Experimental design

The aim of the experimental design was to show any possible influence of key raw material variables on pellet quality parameters. Variables under study were tree species, bark presence and wood fraction: logs with diameters at breast height of 15-20 cm (DBH15), logs with diameters at breast height (DBH) of more than 20 cm (DBH20) or branches. Moreover, the possible interdependence observed among the most important pellet quality parameters was studied through regression analyses.

In parallel to this, mixes of non-debarked and debarked woodchip material within the same species were elaborated in defined percentages. Pellets were produced from this material in order to determine the quantity of non-debarked material that a debarked woodchip assortment can contain to still comply with A1 standard quality following EN 14961-2.

10 different woodchip assortments of 10 kg each one were obtained from 30 trees per each tree species. Woodchips were classified according to the fraction from which they proceeded (DBH15, DBH20 and branches) and according to the bark presence, i.e. barked or debarked logs. A sample of 5 kg of pellets per each woodchip assortment were used for the qualitative analysis in the lab.

3.3. Pellet manufacturing

Pellet manufacturing was performed in AIDIMA experimental pilot plant. Woodchips were received at 40-60% moisture content and stored in ventilated plastic containers (PLASTIPON) for their natural drying. After three weeks and to ensure a sufficient and homogenous moisture content in all samples, woodchips were disposed in a climatic chamber ACS50 (UC50 model, 20kW) at 20°C and 90% of air humidity until 15% of moisture content. After this, woodchips were comminuted to 5mm sawdust with a hammer chipper (OLIGOTECHNOLOGY) in order to obtain homogeneous and comparable particles from all samples. A second reduction of material size was done by a refining mill (MECAFA) to obtain sawdust of 3mm required as input by the pellet machine. A KAHL press with plane matrix of 50kg/h capacity with 3mm circular openings for particle screening was utilized for pelletizing, working at a temperature comprised between 90-105 °C (figure 2a). Pellets of 6mm diameter and 40mm length were produced without using any additive. After its production, pellets were air cooled in uncovered plastic trays in normal climatic conditions of 20 °C and 50% air humidity.

...Figures 2a and 2b about here...

3.4 Pellet analysis

After pellet production, wood pellets were analysed according to the standard EN 14961-2 for non-industrial pellet quality classification. Following quality parameters were analysed:

a) Origin, source and dimensions

The main parameters of the raw material composition of the biomass obtained from the harvesting of *Pinus halepensis* and *Pinus pinaster* have been studied following EN 14961-1 and EN 16127. Moreover, percentage of bark content was measured in non-debarked log samples.

b) Moisture content

The moisture content has been measured following EN 14774-1 with a CARBOLITE stove (model AAF1100).

c) Ash content

The ash content has been analysed following EN 14775 with the same stove. To know ash content in pellets is very relevant for the correct performance of stoves and boilers in which pellets are combusted due to slags and residue depositions (Öhman et al. 2004). On the other hand, a high ash content can indicate the presence of external materials in raw material for pellet production such as additives, dirty, non-wood residues etc. (Francescato et al. 2008). Moreover, a high ash content reduces net calorific value and implies sinterization risk affecting negatively to mill equipment and pellet presses in the industrial manufacturing processes (Lehtikangas 2001).

d) Fine particle content

Fine particles or sawdust has been analysed following EN 15149-2 with a CISA mechanical vibration equipment (model RO200N, 0,45 kW). The fine particle content is a result of the production, packaging and transport processes (Pellets@las 2009). This is a key quality parameter as a limiting factor for pellet movement during boilers feeding. High fine particle content contributes to an inefficient feeding with a more heterogeneous material, which derives in a poor combustion quality (Lehtikangas 2001). An excessive fine particle presence may also cause fire hazards in storage silos and healthy risk for the respiratory tract of the factory operators (Vinterback 2004).

e) Mechanical durability

The mechanical durability has been tested following EN 15210-1 with a pellet durability tester Bioenergy TUMBLER Series (figure 2b). This is a key property for the pellet consumers (Pellets@las 2009) and the only physical metrical parameter considered by industry (Wilson 2010). A high pellet durability is very important to reduce fine particle

1 presence and to maintain a good pellet quality during storage and transport (Filbakk et al.
2 2011). According to Pellets@las (2009), mechanical durability is determined, among other
3 factors, by lignin and water content as well as with compression pressure during pellet
4 pressing and an appropriate cooling process.
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7 *f) Bulk density*
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9 Bulk density has been measured following the instructions of EN 15103. Arshadi et al.
10 (2008) affirm that bulk density depends on raw material and pellet manufacturing
11 conditions such as press pressure, particle moisture content during the pressing, etc. Bulk
12 density is also an important property for transport efficiency (Lehtikangas 2001).
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16 *g) Net calorific value*
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18 Net calorific value has been tested following EN 14918 with calorimetric bomb PARR,
19 (model 1351).
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23 *h) Chlorine and sulphur content*
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25 The chlorine and sulphur content has been determined also with the calorimetric bomb
26 following EN 15289.
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4. Results

4.1 Pellets quality properties

All woodchips assortments were pelletized under the same laboratory conditions. Table 1 shows the average results for quality properties of each woodchip assortment and Table 2 the statistical result of the influence of raw material variables over the quality properties.

...Table 1 about here...

a) Bark content

The obtained results show a higher bark percentage in *Pinus pinaster* (11,43%) than in *Pinus halepensis* (8,76%). Moreover, at the same tree age, a higher diametric class is related with higher bark percentage for both species. The statistical analysis shows no significant influence of the tree fraction on the bark content in the samples performed and studied. Similar results found Bradfield and Michal (1984) for southern softwoods in the USA.

b) Moisture content

The highest moisture content is found for the samples of *Pinus pinaster* barked DBH 20 (6,33%) and the lowest for the samples of *Pinus halepensis* branches (3,33%). The results show a significant higher moisture content in pellet samples with bark presence at the same diametric class. So, at a diametric class of 20 cm, *Pinus halepensis* debarked samples are drier (4,67%) than samples with bark (5,50%) and *Pinus pinaster* debarked samples are also drier (3,83%) than samples with bark (5,83%). This trend coincides with Filbakk et al. (2011), who found pellets performed with *Pinus sylvestris* to contain higher moisture with bark than without bark. Furthermore, the statistical analysis demonstrates the significant influence of bark presence on moisture content of the analysed pellets.

...Table 2 about here...

c) Ash content

The obtained results show a significantly higher ash content in *Pinus halepensis* pellets (in average 0,75%) than in *Pinus pinaster* pellets (0,45%). Additionally, pellets from raw material with bark presence show higher ash percentage (in average 0,60%) than pellets from debarked raw material (0,45%). Moreover, the ash content of pellet samples manufactured from branches is higher (in average 0,90%) than the content observed in pellets from logs (0,53%). Thus, according to the statistical analysis, the ash content is significantly influenced by the bark content for both species. Similar results found Jirjis et al. (2006) for northern European softwoods.

d) Fine particle content

Pinus halepensis samples show an average fine particle content of 0,5%. This value is very similar to the fine particle content of *Pinus pinaster* samples (0,4%). The statistical

1 analysis shows no significant differences in the fine particle content of the samples
2 studied. Similar results found Relova et al. (2009) for pellets manufactured with *Pinus*
3 *caribaea*.
4

5 *e) Mechanical durability* 6

7 The results obtained show slight differences for the mechanical durability between *Pinus*
8 *halepensis* pellets (in average 94,4%) and *Pinus pinaster* pellets (95,9%). Nevertheless, the
9 statistical analysis demonstrates that this difference is significant. Lehtikangas (2001)
10 affirms in this sense that the difference in lignin content between tree species determines
11 wood viscoelastic properties and therefore can influence on mechanical behaviour of
12 pellets. Also Rhen et al. (2005) and Lestander et al. (2012) conclude that mechanical
13 durability, but also bulk density and ash content highly depend on the pelletized tree
14 species.
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17 *f) Bulk density* 18

19 The small differences observed between the bulk density of *Pinus halepensis* pellets (603,3
20 kg/cm³) and *Pinus pinaster* pellets (631,7 kg/cm³) can be considered as significant after
21 the statistical analysis. On the other side, neither the tree fraction (log or branches) nor
22 the bark presence have significant influence on the bulk density. These results coincide
23 with Lestander et al. (2012), who demonstrate that bulk density depend mainly on the
24 wood species.
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28 *g) Net calorific value* 29

30 The net calorific value in dry basis at a constant pressure is very similar for both species,
31 being 17,80 MJ/kg in average (17,84 MJ/kg for *Pinus halepensis* diameter class 20cm with
32 bark and 17,77 MJ/kg for *Pinus pinaster* diameter class 20cm with bark). The statistical
33 analysis shows no significant differences between species. Francescato et al. (2008) also
34 found that the net calorific value is very similar among softwood species in Europe.
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38 *h) Chlorine and sulphur content* 39

40 The results obtained for chlorine (in average 0,02%) and sulphur (in average 0,01%)
41 content show no significant differences between the two pine species analysed.
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45 **4.2 Pellet quality** 46

47 According to the pellet analysis results showed in Table 1, most of the pellets
48 manufactured with woodchip pellets comply with A1 quality requirements following EN
49 14961-2. Exceptions are the pellets manufactured with small dimensions of *Pinus*
50 *halepensis* with bark (PH B 15 and PH B 20) as well as the pellets manufactured with
51 branch material from both species (PH B and PP B), which have a high content on ashes
52 (A>0,7%) reaching only A2 quality.
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56 **4.3 Interdependence between pellet quality factors** 57

58 The summary of regression coefficients given in Table 3 and the interdependence
59 diagnosis show that there exists a highly significant invers linear correlation between bark
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1 percentage present in raw material and fine particle content of manufactured pellets
2 ($R^2=0,981$ for *Pinus halepensis*), as shown in Figure 3. This result coincides with Filbakk et
3 al. (2011), who confirm that fine particles content decreases with the increment of bark
4 content in different samples of pellets manufactured with *Pinus sylvestris*.

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7 ...Figure 3 about here...

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10 A direct correlation can be observed between bark and ash content, but the significant
11 dependence of ash content on the tree species and the lack of enough bark content data
12 per species made no possible to determine a significant regression coefficient.

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15 ...Table 3 about here...

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18 Furthermore, a highly significant positive non-linear correlation can be observed between
19 bark content and mechanical durability ($R^2=0,955$), as shown in Figure 4. This confirms
20 that an increase of bark content increases also mechanical durability. This can be
21 explained according to Lehtikangas (2001) due to that higher lignin and extractives
22 concentration, as occurs in bark in comparison with wood, having a positive effect on
23 cohesion mechanism during pellet manufacturing (Francescato et al. 2008).

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31 Finally, an inverse correlation can be observed between mechanical durability and fine
32 particle content when it is studied separately per pelletized tree species, being higher for
33 *Pinus halepensis* ($R^2=0,980$) than for *Pinus pinaster* ($R^2=0,914$). The interdependence
34 among the rest of pellet properties show no significant correlation values ($R^2 \leq 0,8$).

35 36 37 38 **4.4. Maximum tolerated bark content**

39 One practical result of this research is the possibility to add up to certain percentages of
40 non-debarked forest material (from non-debarked small-sized logs or even biomass
41 residues such as branches) to woodchips obtained from debarked logs in order to still
42 reach high quality pellets with allowable percentages of ash content ($A \leq 0,7\%$), which is
43 the more restrictive quality parameter. In this sense, Table 4 shows that depending on the
44 tree species, it is possible to add up a considerable amount of low value chips from
45 branches or non-debarked small-size logs to high qualitative woodchips from debarked
46 logs. So, for example for *Pinus halepensis* up to 50% of low valued chips from branches
47 could be mixed with 50% of high valued chips from debarked logs with a minimum
48 diameter of 20cm in order to obtain high quality pellets.

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53 ...Table 4 about here...

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56 On the other side, for *Pinus pinaster* up to 65% of low valued chips from branches could be
57 mixed with 50% of high valued chips from debarked logs with a minimum diameter of
58 20cm in order to obtain also high quality pellets.

The rest of quality parameters (bulk density, moisture content and mechanical durability) are not considered critical for obtaining high quality wood pellets because, despite some of them depend on the raw material composition, they can be highly improved with the improvement of adequated pellet manufacturing processes.

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4. Conclusions

1 The research has been focussed on two important Mediterranean pine species: *Pinus*
2 *halepensis* and *Pinus pinaster*. As first conclusion of the results obtained, the pinetree
3 species to be pelletized and the percentage of bark content are the main raw material
4 variables that affect pellet quality parameters. Ash content, mechanical durability and bulk
5 density are mainly affected by the tree species, while bark presence mainly influences
6 moisture and ash content.
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9 Even pellets manufactured from logging residues of *Pinus halepensis* and *Pinus pinaster*
10 (small-sized logs of clearings and crown material such as branches) are able to reach high
11 pellet qualities, if the ash content is duly controlled. A higher bark content in the raw
12 material mixture for pellet manufacturing decreases the percentage of fine particle
13 content, while ensures at the same time higher mechanical durability. All this enables the
14 possibility to include full-tree harvested and chipped material for direct pellet
15 manufacturing. This opens an interesting possibility to reduce production costs by
16 eliminating debarking processes in the forest or in the mill. Nevertheless, it has to be taken
17 into account that *Pinus halepensis* is more sensitive in terms of ash content than *Pinus*
18 *pinaster*.
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21 Finally, the research demonstrates that there is a high potential of utilization of residual
22 forest biomass in the Mediterranean basin for obtaining high quality solid wood biofuels.
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Table 1: Pellet properties and quality classification according to EN 14961-2

| Sample | BC(%) | M (%) | A (%) | DU (%) | F(%) | BD (kg/m ³) | Q (MJ/kg) | Cl(%) | S (%) | QUALITY CLASS |
|----------|-------|-------|-------|--------|------|----------------------------|--------------|-------|-------|------------------|
| PH B 15 | 4,60 | 5,50 | 0,83 | 93,61 | 0,70 | 652 | 17,39 | 0,02 | 0,01 | A2 |
| PH DB 15 | - | 4,83 | 0,62 | 97,11 | - | 618 | 17,38 | 0,02 | 0,01 | A1 |
| PH B 20 | 12,92 | 5,50 | 0,94 | 96,80 | 0,21 | 590 | 17,84 | 0,02 | 0,01 | A2 |
| PH DB 20 | - | 4,67 | 0,55 | 89,79 | 0,29 | 593 | 17,85 | 0,01 | 0,01 | A1 |
| PP B 15 | 9,34 | 6,33 | 0,41 | 94,70 | 0,38 | 638 | 17,32 | 0,01 | 0,01 | A1 |
| PP DB 15 | - | 5,17 | 0,25 | 98,25 | 0,17 | 650 | 17,41 | 0,01 | 0,01 | A1 |
| PP B 20 | 13,53 | 5,83 | 0,46 | 98,60 | 0,21 | 629 | 17,77 | 0,02 | 0,02 | A1 |
| PP DB 20 | - | 3,83 | 0,20 | 94,61 | 0,63 | 600 | 18,65 | 0,02 | 0,02 | A1 |
| PH BR | - | 3,33 | 0,85 | 79,31 | 0,76 | 563 | 18,36 | 0,02 | 0,02 | A2 |
| PP BR | - | 5,83 | 0,94 | 93,55 | 0,71 | 640 | 17,69 | 0,01 | 0,01 | A2 |

PH= *Pinus halepensis*, PP= *Pinus pinaster*, B= Barked log, DB= Debarked log, 15= DBH 15 cm, 20= DBH 20cm, BR= Branches, BC=bark content, M=moisture, A=ash, DU= durability, F= fines, BD= bulk density, Q= net calorific value, Cl= Chlorine, S= sulphur

Table 2: Influence of raw material variables on pellet properties

| P-value | BC(%) | M (%) | A (%) | DU (%) | F(%) | BD (kg/m³) | Q (MJ/kg) | Cl(%) | S (%) |
|----------------|--------------|--------------|--------------|---------------|-------------|----------------------------------|----------------------|--------------|--------------|
| Specie | (n.s.) | (n.s.) | 0,0004 | 0,0492 | (n.s.) | 0,0310 | (n.s.) | (n.s.) | (n.s.) |
| Tree fraction | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) |
| Bark presence | - | 0,0308 | 0,0023 | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) | (n.s.) |

n.s. = not significant.

Table 3: Summary of regression coefficients for interdependence of properties studied

| R² | BC(%) | F(%) | A (%) |
|----------------------|--------------|--------------------------|--------------|
| BC (%) | - | 0,981 | * |
| DU (%) | 0,955 | 0,980 (PH) 0,914 (PP) | - |

* only for *Pinus pinaster*

Table 4: Summary of possible sample mixtures of raw material with bark that allow the production of A1 quality pellets

| Specie | Sample mix | %DB | %B |
|---------------|-------------------|------------|-----------|
| PH | DB 15 - B 15 | 65 | 35 |
| | DB 20 - B 20 | 65 | 35 |
| | DB 15 - BR | 70 | 30 |
| | DB 20 - BR | 50 | 50 |
| PP | DB 15 - BR | 35 | 65 |
| | DB 20 - BR | 35 | 65 |
| | B 15 -BR | 55 | 45 |
| | B 20 -BR | 50 | 50 |



Figure 1a: Selected logs of *Pinus halepensis* with diameters at breast height (DBH) of more than 20 cm (DBH20)



Figure 1b: Selected material for chipping and pelletizing of *Pinus pinaster* with diameters at breast height (DBH) of 15-20 cm (DBH15)



Figure 2a: Sample of experimental pellets manufactured with KAHL press with plane matrix of 50kg/h capacity



Figure 2b: Test of mechanical durability with Bioenergy TUMBLER Series following EN 15210-1

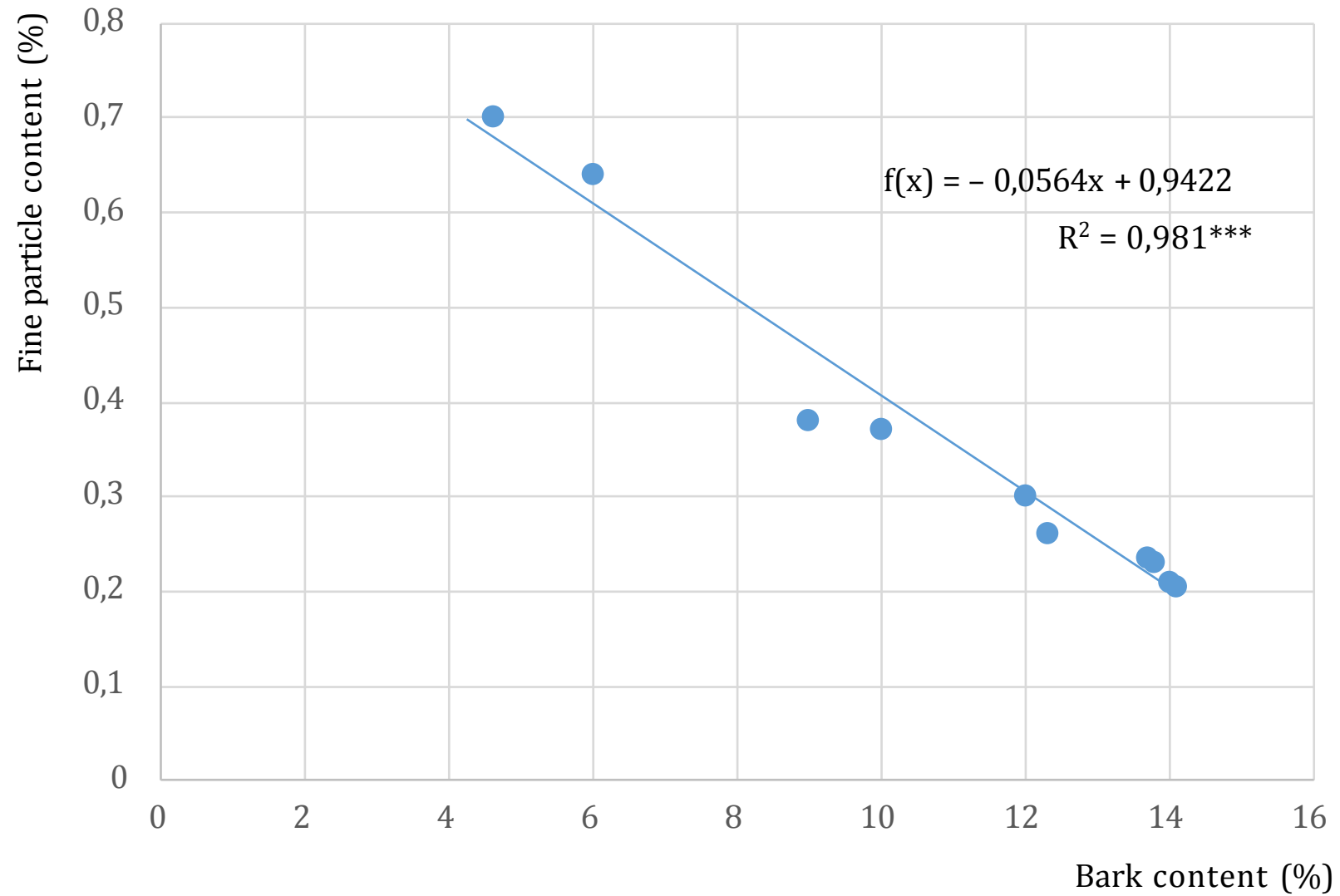


Figure 3: Regression analysis between bark percentage present in raw material and fine particle content of manufactured pellets

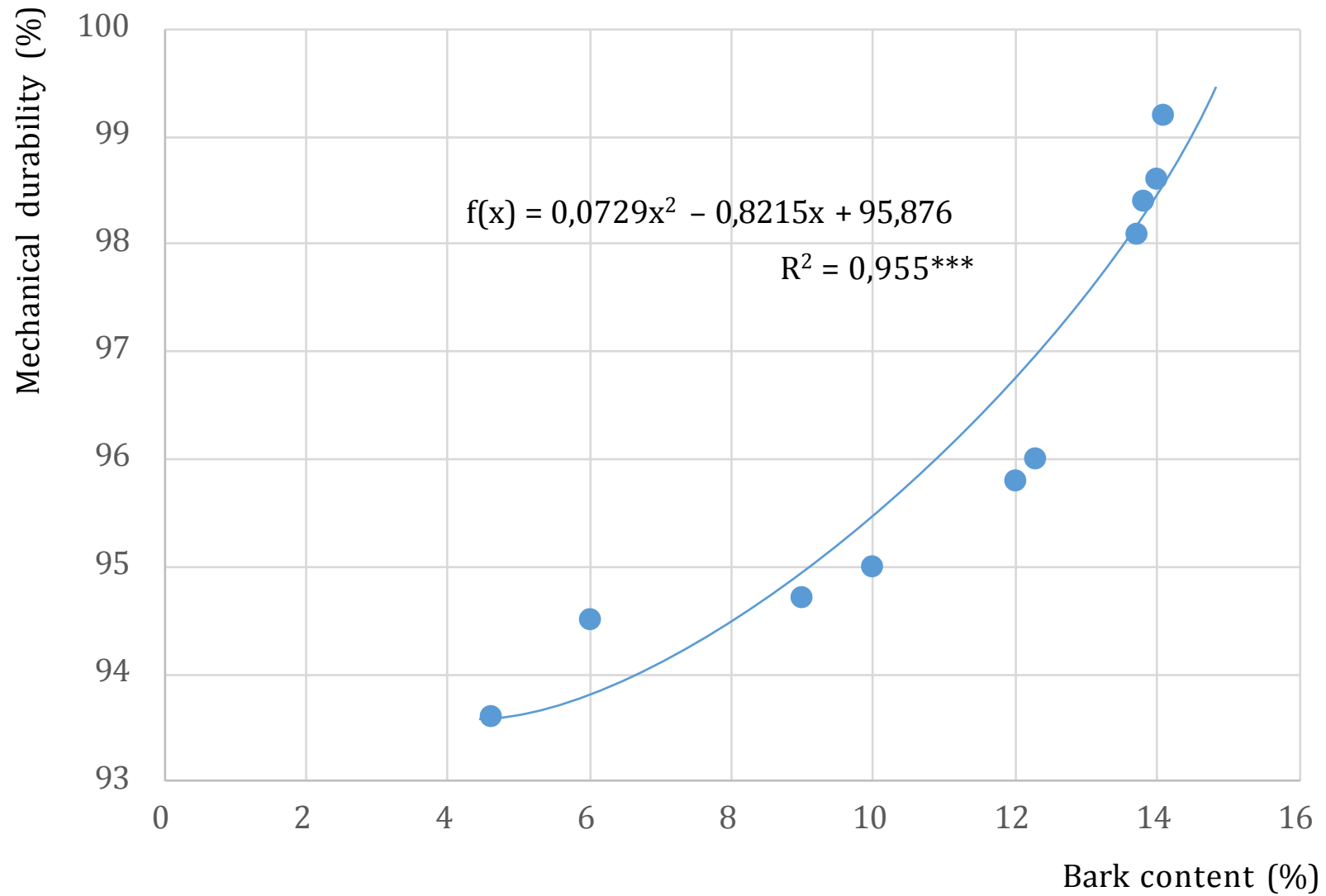


Figure 2: Regression analysis between correlation bark content and mechanical durability of manufactured pellets