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THE NEW PORTUGUESE ENERGY CHALLENGE? PELLETS FROM SHRUBS.

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Abstract. *The large presence of Cistus (rock-rose) and Cytisus spp (broom) in 64% of the Portuguese mainland is a potential source of an autochthonous renewable energy form. It enables the eventual development of a strategy for sustained exploitation. Beyond capturing* CO_2*, the benefits resulting from its use as fuel will bring additional, environmental, economic and social gains. The perception of the presence of such economic value in vast areas so far unsuitable for agriculture will contribute to the necessary countryside repopulation; together with this important benefit, there is the decrease in energy imports, in a country where three quarters of the energy spent is produced abroad. As, in addition, two thirds of the final energy is consumed in heating and cooling, recent developments in biomass combustion efficiency and handling through automated boilers and stoves, was supported, mostly, by means of densification of this kind of lignocellulosic materials into a cylindrical shape. To determine the potential of the Cytisus and Cistus species, 1 and 1.5 mm mean diameter particles were used to produce 10 mm diameter pellets. Moisture contents in the range between 8 and 15% were used and the pellets were fabricated under laboratory and industrial conditions. Using a device coupled to an instrumented universal testing machine, and using pressures between 75 to 125 MPa, the specific energy was calculated for each case using three compression velocities (0.5, 1.0 and 1.5 MPa/s). Here are presented the results of submitting those products to tests, as well as of the comparison with the pellets of the same materials that were produced with industrial machinery, in terms of: (a) stability in contact with moisture and (b) durability under conditions of mechanical wear.*

Keywords*: bioenergy, pelletizing, broom and rock-rose.*

1. INTRODUCTION

Portugal imports 83.3% of the primary energy consumed and only a third of the final energy consumption is used in transport, the remaining two thirds are used to lighting, heating and cooling. The contribution of Renewable Energy Sources (RES) for the primary energy consumption in 2008 was 17.6%. The shortage of indigenous energy resources, oil in particular, has motivated the pursuit of solutions to minimize the national energy bill. The increase of installed power from RES in 2008 reached a value of 8458 MW and generated 15.4 GWh. The 3030 MW of wind power and 4857 MW of hydropower generated 13045 GWh, meaning that 84.6% of all the renewable power came from renewables. Biomass accounts only for 6% (492 MW) of the total installed RES base power and generated 2128 GWh, 13.8% of the global electricity generated based on RES (DGEG, 2010). The overall goal for the Portuguese share of energy from renewable sources in gross final consumption of energy in 2020 is 31%, as established by the European Union (EPC, 2009).

Biomass will become an important and sustainable contributor to the global energy production. Though still more expensive than fossil fuels, trends show that an efficient cycle that includes transport, harvest and combustion, can make it economically competitive (Khan et al., 2009). The potential contribution of bioenergy to world energy demands considers three categories of resource biomass: forest or agricultural residues; various organic waste, dedicated biomass production on lands of different types: the production of forage or pasture, plantation forest and the low productivity of marginal and degraded lands, (El Bassam, 2010).

38% of the Portuguese mainland is forest, 3.3 Mha of tree stands of soft- and hardwood, led by *Pinus pinaster Aiton*, *Eucalyptus globulus Labill* and *Quercus suber*. Farms occupy 33% of the country (*i.e.*, 2.8 Mha) and 26% is unproductive (IFN, 2007). With the revision of national forest inventory of 2005 there is still a predominance of understory vegetation species of the genus *Cytisus* in 4.3% of forest area, or 145 kha, and 7.5% of genus of *Cistus*, or 250 kha (Godinho *et al*., 2005).

Bioenergy culture depends on land availability, given the need to ensure the growing global demand for food, combined with environmental protection, sustainable land management, water supply and other sustainability requirements, such as maintenance of biodiversity (El Bassam, 2010).

The abundant presence of shrubs *Cistus* (rockrose) and *Cytisus spp* (broom) in 64% of the Portuguese mainland is a latent source of native renewable bioenergy. Without the forest, Portugal has only 2.3 Mha of uncultivated, marginal and abandoned land, the opportunity to exploit energy-dedicated native shrub infesting species, easily installed and adapted to proliferate and survive incendiary devastation and summer droughts are: a) *Fabaceae*, legumes of the genera *Cytisus* and *Retama* like *C. multiflorus*, *scoparius*, *striatus* and *Retama sphaerocarpa* (brooms), which prevent erosion, restore, re-qualify and fertilize the soil with their ability to fix nitrogen (Fabião and Pereira, 1979; Abreu *et al.*, 2007), medicinal and melliferous (Carvalho *et. al.*, 2001), producer of high quality and easy growing biomass (El Bassam, 2010), once used as natural fertilizers in traditional cereals (Estabrook, 2006; Estabrook, 2008); b) *Cistaceae*, as the rockrose, *Cistus ladanifer xerophilous Sclerophyllous* shrub, which grows easily in dry, rocky, degraded soils, with high ability to photosynthesize and fix carbon (Simões, 2003). Whose bio-ecology and production yield was indicated to be incorporated into a sustainable rural development (Borges and Almeida, 1996, Borges *et al.*, 1992). In addition to phyto-stabilization and soil minerals recovery (Abreu *et al.*, 2009), it allows for bioethanol production (Ferreira *et al.*, 2009).

Job creation, agro-forestry enhancement, population establishment in abandoned areas and the reduction of the imported energy bill encourages the use of native bioenergy. Bioenergy based energy conversion systems are the only renewable source, besides hydro-energy, to suit different needs, likely to produce, electricity or heat only and when needed (Bain *et al.*, 1996).

Despite the technological constraints of biomass combustion, and the associated environmental and economic impacts, they are surmountable (Khan *et al.*, 2009). Combustion of lignocellulosic materials requires less air, compared with other solid fuels. A homogeneous mixture of air fuel is virtually impossible to obtain with biofuels, so that the densification is a way to improve their performance in combustion (Tabarés *et al.*, 2000).

The technology enabled the densification of biomass to bioenergy expansion as a product. Beyond improving its volumetric heating value, it facilitates handling, storage and transportation. It consists basically on the compression of solid particles in a confined volume, either with low pressure using additives and binders, or with medium pressure and heating, or by a simple application of high pressure (Grover and Mishra, 1996).

In Europe, the densification process has tended to pelleting, compaction of particles by means of rollers, on a rotating perforated cylindrical matrix. From that it results homogeneous cylinders of the same diameter of the perforations of the matrix (6, 8, 10 mm) and length of less than 4cm. The pellets production quality is principally dependent upon the characteristics of the base material (Jenkins, 2010).

In evaluating the efficiency of converting biomass to bioenergy, it is essential to consider the intrinsic nature of its chemical and physical properties (density, water content, calorific value, cellulose lignin ratio, percentage of carbon, volatile substances, ash content, metallic and alkaline content), which influence the parameters of conversion processes (Mckendry, 2002).

The use of pellets of high density, low water content, size and homogeneous composition, can reduce or completely avoid the difficulties of using conventional biofuels. Such high quality biofuel allowed the development of more efficient combustion technologies. Stoves and boilers with higher yield, cleaner combustion, with external furnace using inserted and integrated automatic ignition, with automatic feeding systems, lead to an operational comfort similar to that found with basic fossil fuel appliances (Obernberger and Thek, 2010).

The availability of raw material for pellets production is increasingly a problem due to market growth. Therefore, herbaceous or lignocellulosic energy crops gain greater importance when low-cost materials such as sawdust, wood chips and tape, begin to scarce (Obernberger and Thek, 2010). There is still a long way to transpose in the production of pellets. The production from an unknown raw material is not for anyone! Even companies that operate for some years now they have difficulties and conclude that not all materials are likely to be pelleted at least in terms of immediate knowledge (Holm *et al*., 2006).

However, there are factors to be studied, due to their influence on the pelletization and subsequent combustion, capable of determining the future good or bad performance of such product. There are unanswered questions such as: the need to use a new technique for straw defibrillation to increase its durability, influence of the post-production cooling temperature of the pellet, stabilization time after the pellet fabrication process and its influence on mechanical strength and hygroscopic, the addition of hydrogen peroxide to increase the yield of pellet while decreasing the calorific value of the pellets, the addition of binders to benefit from cohesion and consequent durability of pellets, the pH of the soil from which the biomass is original, a parameter that has strong influence on the melting temperature of ash (Obernberger and Thek, 2010).

Inserted in a broader work of evaluating the potential of native species for bioenergy, it was intended with this work the production in industrial equipment of *Cytisus multiflorus* and *Cistus ladanifer* pellets. For this purpose, and using an Anyang General International industrial pellet press model GC-9PK200, pellets were produced of the genera *Cistus*, *Cytisus*, well as mixtures of the latter with sawdust of *Pinus* and *Fagus* tape left over from the typical wood industries. It was determined the durability, density, percentage of fines and stability of pellets against their moisture content. The percentage of ash content of the pellets was also determined.

For comparison with the industrially produced pellets, laboratory made pellets with 10 mm diameter of *Cistus* and *Cytisus* were also manufactured. To this end, a piston device chamber adapted to a universal testing machine (Marques *et al*., 2009), was used and the energy of densification was determined for compression rates in the 0.5-1.5 MPa/s range. Pellets of mixtures of *Cistus* and *Cytisus* with 25, 50 and 75% (wt) were produced to evaluate the potential of *Cistus* as a natural aggregative additive.

2. MATERIAL AND EXPERIMENTS

2.1. Material preparation

For the present work, *Cytisus multiflorus*, *Cytisus striatus* (brooms) and *Cistus ladanifer* (rockrose) were harvested in a forest situated at the center of Portugal. Two batches of *Cytisus* were prepared for the drying process. One batch, covered and protected from the weather, was dried in air. Only a shed covered by a zinc roof, branches, leaves and flowers were piled in the soil, as collected, with a height of 1m. This batch, thast dried during 90 days during autumn and winter, was kept stored until milled. The second batch, as well as the *Cistus ladanifer*, was dried in a solar greenhouse located at the ESTGV roof. The material, after drying, was ground in two separate equipments. All materials used in the laboratory were previously cut into small pieces on a band saw EBS ELU-306 and subsequently milled in a knife mill with a P19 Fritsh 2 mm sieve. The raw material for use in industrial equipment was ground in a hammer mill TFS420 Genco. The remainder biomass, *i.e.*, the timber sawdust from pine and beech tape, was supplied by one sawmill of the region.

2.1.1 Chemical characterization of *Cytisus* and *Cistus*

Among the chemical components of biomass constituents, those who stand out more positively in their densification are lignin and protein substances. The presence of lignin in the raw material increases the binder characteristics of the particles due to its low melting point of 140° C (Mani *et al*., 2006).

An immediate analysis of the species collected was made by LNEG-National Laboratory of Engineering and Geology, whose information is summarized in Tab. 1. In this analysis, in addition to quantifying the chemical constituents of the species studied, HHV and LHV values were determined.

Table 1. Proximate analysis of *Cytisus multiflorus* (broom) and *Cistus ladanifer* (rockrose).

		***Cytisus* multiflorus (broom)**			***Cistus* ladanifer (rockrose)**		
Properties	**Methodology**	**Unit**	**As received**	**Dry basis**	**Unit**	**As received**	**Dry basis**
Total Moisture	CEN/TS 14774-2	% (m/m)	8.3	---	% (m/m)	26.8	----
Ash	CEN/TS 14775	% (m/m)	1.2	1.4	% (m/m)	1.8	2.4
Volatile	CEN/TS 15148	% (m/m)	74.9	81.6	% (m/m)	57.2	78.1
Fixed C	ASTM D 3172	% (m/m)	15.6	17.0	% (m/m)	14.2	19.5
Total C	CEN/TS 15104	% (m/m)	46.0	50.2	% (m/m)	36.2	49.5
Total H	CEN/TS 15104	% (m/m)	6.4	6.0	% (m/m)	7.1	5.4
N	CEN/TS 15104	% (m/m)	1.1	1.1	% (m/m)	0.6	0.8
S	CEN/TS 15289	% (m/m)	<LQ=0.6	<LQ=0.6	% (m/m)	1.76	2.41
HHV	CEN/TS 14918	kJ/kg	18 144	19 786	kJ/kg	15 684	21 427
LHV	CEN/TS 14918	kJ/kg	16 756	18 491	kJ/kg	14 164	20 233

Table 2 summarizes the chemical composition of *Cistus ladanifer* and *Cytisus striatus*. A significant difference in the percentage of total lignin between *Cistus* and *Cytisus* can be detected.

Table 2. Chemical composition of *Cistus ladanifer* (rock-rose) and *Cytisus striatus* (broom), adapted from Gil *et al.* (2009).

Species / origin		**Extractives (%)**	**Ash (%)**	**Insoluble lignin(%)**	**Soluble lignin (%)**	**Total lignin (%)**	**Glucose (%)**	**Xylose (%)**	**Total sugars (%)**
Rock-rose *Cistus*	Stalks Without bark	1.8	0.8	23.0	1.9	24.9	-	-	-
	Whole plant	7.4	3.1	32.0	2.2	34.2	33.1	15.9	49.0
Broom *Cytisus*	Stalks Without bark	3.4	0.6	22.0	2.1	24.1	-	-	-
	Whole plant	4.7	0.8	22.4	2.3	24.7	45.9	19.5	65.4

2.1.2. Average particle size of raw material

The particle size influences the durability of pellets; the smaller the particle size, the greater the pellets durability. Fine particles generally hold more moisture than larger ones, allowing a greater degree of packing. The larger cracks are weak spots and, therefore, the pellets are more prone to fragmentation. Pellets of good durability should be obtained from an average size between 0.5 and 0.7 mm, which usually appears as a breaking point from the above dimensions of 1.0 mm (Kaliyan *et al.*, 2009).

The size distribution and the average particle size (*dwg*) of all materials were determined according to ISO 3310-2 1999-01. In accordance with the provenance of grinding, Tab. 4, all material was sieved for 10 minutes with a 1.50 mm amplitude in a "RETSCH" AS200 sieving device, following the gravimetric method (Hartmann *et al.*, 2006). In this case, it was possible to observe the differences between the two grinding equipments that were used, as illustrated for the rockrose, in Fig.1.

For the manufacture of pellets in the laboratory, broom and rockrose ground in the mill of knives, with average diameters of 1439 and 1090 µm, respectively, were selected and packaged. In the industrial extrusion of pellets, broom and rockrose with average diameters of 918 and 1088 µm, respectively, from the hammer mill industry were used. The sawdust of pine and beech tape were used as received from the sawmill.

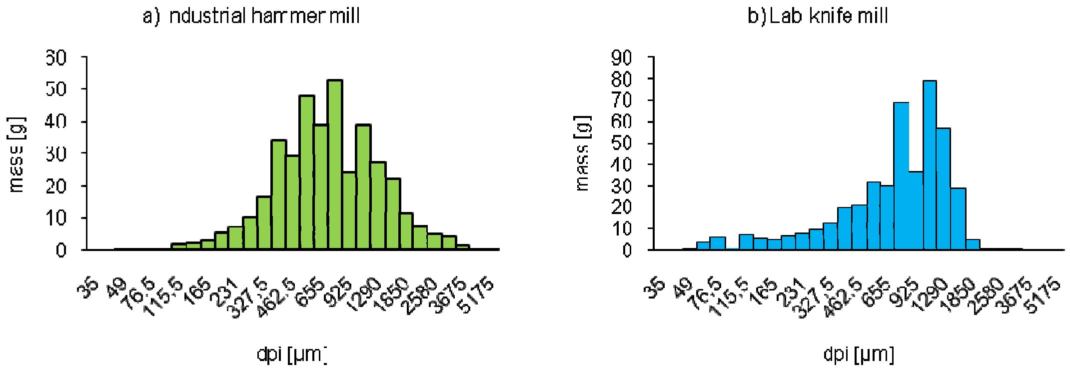

Figure 1. Differences in the distribution of particles, between grinding mills. a) Milled rockrose with a mean diameter of 857 µm; b) Milled rockrose with a mean diameter of 833 µm.

2.1.3. Moisture content of raw material

The raw material, and the pellets produced, were dried in a convection oven (Binder SDL115), at a temperature of 105 ± 3°C, until constant mass was reached (Samuelsson *et al.*, 2006). After cooled to room temperature, 21°C, in a desiccator with silica, the water content of the materials was determined according to Eq.s (1) and (2):

$$H_{wb}= [(m_w - m_d)/m_w].100 \tag{1}$$

$$H_{db}= [(m_w - m_d)/m_d].100 \tag{2}$$

wet and dry base respectively. From these calculations, moistures obtained (wt basis) were: rockrose, 11.23-12.65%, broom, 9.55-11.36%, pine sawdust, 37.48-59.95%, and beech, 49.34-97.40%.

2.1.4. Bulk density of raw material

In manufacturing pellets through pure compression, the initial volume of material is essential to assess the success of determining the densification and compaction energy. The knowledge of that volume permits the right piston placement at the beginning of the experiment, without risk of pre-compressing the material. The pellets were put inside 0.5 l flask, and previously milled sample material was poured in the center to reach the top level. The determination of the used mass allows the calculation of the value of mass per unit of volume, *i.e.*, the bulk density of the sample (Lam *et al.*, 2008). The average density value, obtained after three determinations, was adopted and the results were: rockrose 404.2 kg/m^3, broom 240.4 kg/m^3, pine sawdust 267.2 kg/m^3 and beech 227.1 kg/m^3.

2.3. Experimental Work

After coding the base material in order to fit it to the production map of Tab.5, the samples were packed in sealed polyethylene boxes, for transportation and storage, thus ensuring no properties change.

Table 3. Map of the pellets produced.

Pellets code	Milling equipment	Raw material	Origin
P1	Lab knife mill	100% Cytisus	Laboratory manufactured
P2	Lab knife mill	100% Cistus	Laboratory manufactured
P3	Lab knife mill	50% Cytisus and 50% Cistus	Laboratory manufactured
P4	Lab knife mill	75% Cytisus and 25% Cistus	Laboratory manufactured
P5	Lab knife mill	25% Cytisus and 75% Cistus	Laboratory manufactured
P6	Industrial hammer mill	20% Cytisus** and 80% Pinus	Industrially produced
P7	Industrial hammer mill	40% Cytisus** and 60% Pinus	Industrially produced
P8	Industrial hammer mill	50% Cytisus** and 50% Pinus	Industrially produced
P9	Industrial hammer mill	80% Cytisus** and 20% Pinus	Industrially produced
P10	Industrial hammer mill	100% Cytisus**	Industrially produced
P11	Lab knife mill	100% Cytisus	Industrially produced
P12	Industrial hammer mill	100% Cistus	Industrially produced
P13	Industrial hammer mill	20% Cytisus** and 80% Fagus	Industrially produced
P14	Industrial hammer mill	80% Cytisus** and 20% Fagus	Industrially produced
P15	-	Commercial	Industrially produced
P16	Industrial hammer mill	20% Cytisus** and 80% Pinus	Laboratory manufactured
P17	Industrial hammer mill	40% Cytisus** and 60% Pinus	Laboratory manufactured
P18	Industrial hammer mill	50% Cytisus** and 50% Pinus	Laboratory manufactured
P19	Industrial hammer mill	80% Cytisus** and 20% Pinus	Laboratory manufactured
P20	Industrial hammer mill	20% Cytisus** and 80% Fagus	Laboratory manufactured
P21	Industrial hammer mill	80% Cytisus** and 20% Fagus	Laboratory manufactured
P22	Industrial hammer mill	100% Pinus sawdust	Laboratory manufactured
P23	Industrial hammer mill	100% Fagus tape	Laboratory manufactured
P24	Industrial hammer mill	100% Cytisus**	Laboratory manufactured

** (Dry Cytisus under zinc roof)

2.3.1. Compression tests

The pellets P1-P5 and P16-P21 (Tab. 3), with one gram and 10 mm in diameter, were manufactured in a piston-plunger system adapted to a universal testing machine connected to a computer. The initial length of the piston in the chamber was set according to the bulk density of each material; the compression movement took place in two phases, a pre-load of 1 kN and a maximum load of 15.7 kN (200 MPa).

With a compression speed of 1.44 kN/min, the reduction of the voids and the adjustment of the material to the chamber were made until a force of 1 kN was attained. From that point on, the test evolved according to one of three types of programmed loading speed: 7065, 4710, and 2355 kN/min (1.5, 1.0 and 0.5 MPa/s, respectively).

In the end, the pellet was extracted from the chamber, weighed and measured three consecutive times (length and diameter) with a caliper, and the average value recorded.

The information of the force *versus* displacement evolution during the formation of the pellets was saved in the acquisition system and used to calculate the work of compression (W_C). For each subsequent analysis, the compression process was performed five times.

2.3.2. Industrial production of pellets

The pelletizing process, in addition to being a complex interaction between the particles (and their constituents) and the applied forces, also requires some knowledge about the mechanisms of compression, in order to be optimized for a given material. The fuel pellets extrusion is related to the biomass materials used.

Due to its intrinsic random variability, different physical and chemical characteristics, reproducibility of the mass production of pellets for a given material is not acquired. The feasibility of industrial production of pellets of *Cytisus* and *Cistus* was tested, beginning with a base material that would insure pellets formation to take place: pine.

A pellet press of 7.5 kW was used, with a production capacity of 200 kg/h. Previously characterized material was pressed by the rollers through the matrix channels. After twenty minutes running on empty, to allow matrix heating up, sawdust was poured into the hopper inlet. The high moisture content of sawdust made pine sawdust pellets began to leave the window extraction soft and wet.

Eventually, the extruder reached approximate steady state conditions. As broom and dry rockrose were smashed in the hammer mill as collected, some amount of leaves was present, increasing the percentage of protein substances in the raw material and, thereby, improving aggregate performance (Mani *et al.*, 2006).

In correspondence with the proportions of Tab. 5, portions of broom sawdust were weighed in a digital scale. The production of pellets begun by the sequence reproduced in Tab.5: first pellets to be were produced were "P6", 20% *Cytisus* and 80% sawdust. The pellets obtained were stored in large open plastic bags and allowed to cool off.

2.3.3. Pellets characterization methods

For the integration of commercial pellets and *Cytisus* and *Cistus* as a potential solid biofuel, it is necessary to verify the fulfillment of the minimum standard conditions. The pellets obtained from lignocellulosic origin exclusively were qualified by dimensions, density, water content, ash content, bulk density and durability, as shown in European Standards (Alakangas *et al.*, 2006; Alakangas, 2010), whose results are summarized in Tab.4. Additionally, they were given a rating under the standard EN 14961-1, based on those results.

2.3.3.1. Determination of dimensions and particle density

Average diameter and length were calculated after determining the dimensions of 15 randomly selected elements, using a caliper precision 0.1mm (Obernberger *et al.*, 2004). Together with a precision lab scale (0.0001 g resolution), it was possible to apply the method of the stereometric measurement to calculate the pellets density (Rabier *et al.*, 2006). The bases of each cylindrical pellet were previously normalized to ensure its perpendicularity to the longitudinal axis. The diameter of each pellet was measured three times, at the ends and at the middle of the length, and two length measurements were performed, with the second one taking place after making a rotation of the pellet of 90º. The results of these determinations are synthesized in Tab.4.

2.3.3.2. Determination of bulk density

As stated in section 2.1.4., in determining the bulk density of pellets a 500 ml graduated cylinder was used to measure the volume; the weight of the sample was obtained using the lab scale. The sample pellet was gently released into the cylinder until it reached the level corresponding to the pre-established volume. After weighing the mass of the sample, the bulk (or apparent) density was calculated, with three measurements averaged, as summarized in Tab.4.

2.3.3.3. Determination of fine particles in the pellets produced industrially

The majority of solid biofuels is relatively fragile and disintegrates when exposed to friction and impact, giving rise to fine particles. Since the percentage of fines is a qualifying characteristic, the percentage of fines was determined only in industrially produced pellets. Opening the bags of pellets extruded in the pellet press (section 2.3.2.) in the laboratory, it was observed that they contained some very fine particles. To determine the percentage of fines from the pellets, the sample was sieved in the first place with all of the packaged material using a 3.15 mm sieve to separate those fines. The sample was placed in the sieve of 3.15 mm, stirred vigorously ten times sideways and the material that remained inside the 3.15 mm sieve weighed. The pan base was also weighed t with the resulting fine sieve. According to EN 1519-1, this was done for each type of industrial pellets, referred to in Tab.5, using sampling weights of ±725 g. To calculate the percentage of the corresponding fine, Eq. (3) was used and the results presented in Tab.4.

$$\%\ \text{Fines}=[(\text{Weight of Base Pan+Fines})-(\text{Weight of Base Pan})]/(\text{Initial sample weight})\times 100 \tag{3}$$

2.3.3.4. Determination of the pellets moisture content

As is described in section 2.1.3., 10 g of pellets of each sample were dried at 105 ±3°C in a convection oven, until constant mass was reached, following the method described in CEN TC335 Solid Biofuels (Samuelsson *et al.*, 2006).

2.3.3.5. Absorption test

As with textiles and food, the lignocellulosic interacts with the relative humidity of ambient air to reach the equilibrium moisture sorption (absorption, desorption) at constant temperature (Hartley *et al.*, 2008). The conditions for storage and transport to which the pellets are subjected after production influence their final quality. The storage of wood pellets is substantially different from that of unprocessed raw materials. It is known that materials processed hot, at temperatures higher than 100°C, have low moisture content and a low sorption isotherm, which also prevents microorganisms growth (Lehtikangas, 2000; Hartley *et al.*, 2008). However, during storage, handling and transportation, if not tightly packed, pellets are subject to the environment and its hygroscopic state needs evaluation. To analyze the relative humidity, there was a maintenance test of pellets produced in an environment of fixed relative humidity (85 - 90% RH, 21°C) at room temperature. To perform the experiment, two plastic cake boxes of 32 cm in diameter were used. Under the support network with a depth of 1 cm, 0.8 l of water was poured. Using a hygrometer device, the environment is monitored inside the boxes for 5 days. Two pellets of each type and known moisture were weighed before being tested in the saturated environment. They were placed in watch glasses, to prevent direct contact with water and to be easily identifiable. The pellets were weighed daily from 24 to 24 hours. The percentage of moisture absorption was calculated as the ratio between the final mass and initial mass and is presented in Tab.4.

2.3.3.6. Determination of mechanical durability

To describe the physical quality of solid biofuels, the mechanical durability along with the density, are the most relevant properties. The production of fine particles by abrasion during transport and storage of pellets is related both to the distribution and particle size of origin, as with its propensity for aggregation when compressed. The dust emissions resulting from higher or lower susceptibility to abrasion of the pellets, an inconvenience for consumers, is also a health risk and an explosion hazard (Temmerman *et al.*, 2006). With the exception of pellet types P3, P4, P5 and P16-P21, due to insufficient quantity of pellets, samples with ±500 g were used to calculate the mechanical durability. Each sample was weighed and then released into a ASAE container with 300 × 300 × 125 mm^3 (internal) and tumbled at 50 rpm. After 10 minutes at 50 rpm, the sample was sieved manually for 3.15 mm and weighed. The ratio of the mass left in the sieve and the initial sample is defined as the mechanical durability of the pellets and is expressed as a percentage. The durability of pellets P6-P15, the result of three iterations average, is shown in Fig.2.

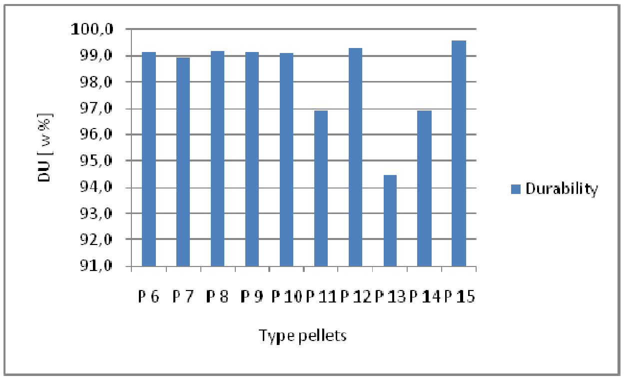

Figure 2. Comparison of the mechanical durability of pellets produced P6-P15 (commercial).

2.3.3.7. Determination of ash content

Establishing the ash content of the pellets produced was done according to ASTM E1755-01, used in a furnace at 575 ±25ºC. Two grams of each type of pellet P1-P15 (Tab. 5, section 2.3.) were weighed together with crucibles of silica in an analytical lab scale, and kept within the oven until constant weight was reached. Simultaneously, the corresponding pellets were dried in a convection oven at 105 ±3°C for moisture assessment, since the samples were air dried (section 2.1. 3.). Once constant weight (± 0.3 mg) was attained, the crucibles were cooled in a desiccator and weighed. After determining the corresponding water content of the pellets tested, the total percentage of solids was calculated, to determine the oven dry weight (ODW) using Eq. (4).

$$\text{ODW} = (\text{Weight air dry sample} \times \% \text{ of Total solids})/100 \qquad (4)$$

$$\% \text{ of Ash content} = [(\text{Weight crucible plus ash} - \text{Weight crucible})/\text{ODW}] \times 100 \qquad (5)$$

With the dry weight of each sample the ash content was calculated through the Eq. (5). The results are summarized in Tab.4.

3. RESULTS AND DISCUSSION

3.1. Compression test results

Figures 3 to 4 correspond to experiments with higher velocity (1.5 MPa/s) of pellets of type P1-P5 (Tab. 3, section 2.3.). The black line represents, in all Fig.s (3-8), the *Cytisus* material (P1), to allow the comparison with the other combinations.

It can be seen in all figures that the *Cytisus* densified more than mixtures with *Cistus*. Figure 3 (left side) compares the curves of work of densification in the formation of pellets made with 100% broom and rockrose. The difference in energy (16 J lower) required by the same densification (70%) between broom and rockrose is considerable.

On the right side of Fig.3, a decrease in the predisposition for the densification of the mixture in relation to substances alone can be noticed.

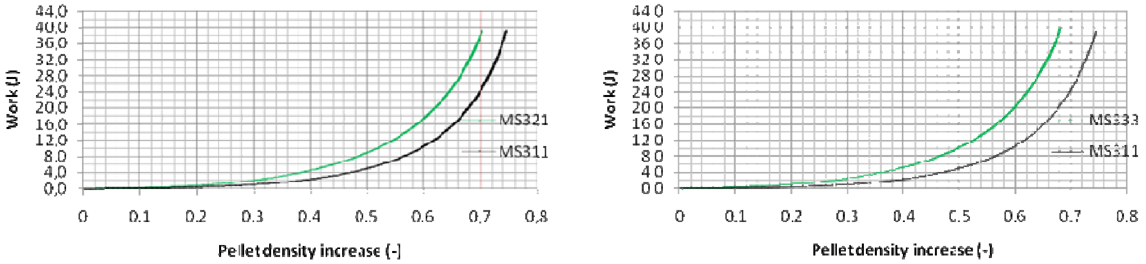

Figure 3. Diagrams of energy spent *vs* densification of 10 mm diameter with a load rate of 1.5 MPa/s. On the left side, P1 P2 (*Cistus* - green line) pellets; on the right side, curves of densification of pellets P1and P3, broom and rockrose (green line), mixed in equal parts.

In Fig.4 (left diagram), with the increase in the percentage of *Cytisus* in the mixture (P4 pellets) the material densified about 70%. It can also be observed that for the densification of 70% an energy difference of 15 J between them remains. The percentage of rockrose in the mixture is 25%. Thus, one would expect that the curves were closer.

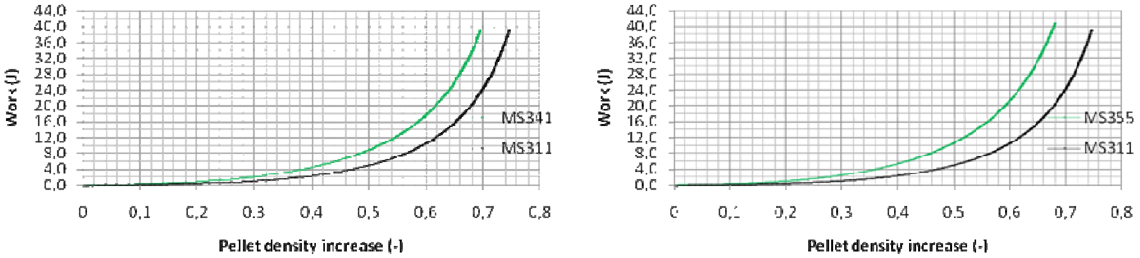

Figure 4. Diagrams of energy spent *vs* densification of 10 mm diameter with a load rate of 1.5 MPa/s. On the left side, P1 P4 (green line) pellets. On the right side, P1 P5 (green line) pellets.

The right diagram of Fig.4 can be compared with the right diagram of Fig.3. The mixture contains 75% of rockrose and, once again, the principle of superposition of effects can be noticed, *i.e.*, small percentage of broom (25%) makes the curve (green line) to move slightly to the left, according to the *Cistus* percentage increase.

The behavior of the mixtures described above remained unchanged for the other two test velocities (0.5 and 1.0 MPa/s). Figure 5 shows the equivalent diagrams of the left side of Fig. 9 for 0.5 and 1.0 MPa/s. Figure 5, left, shows the relation between the energy of densification of the pellet P1 (broom) with P2 (rockrose), but for velocities of 0.5 MPa/s.

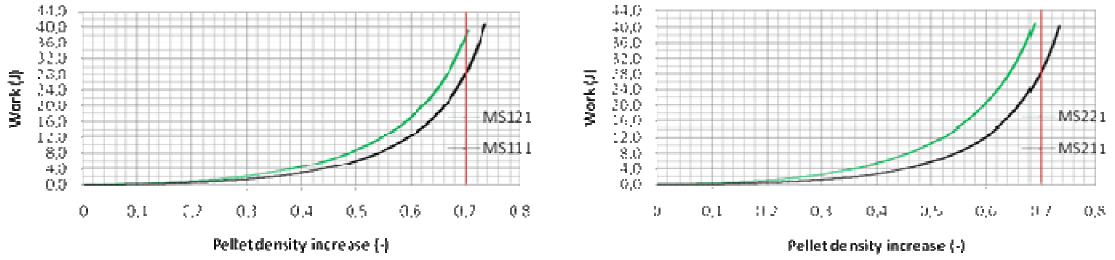

Figure 5. Diagrams of energy spent *vs* densification of 10 mm diameter with a load rate of 0.5 MPa/s on left side and 1.0 MPa/s on the right side. Both diagrams for P1 and P2 pellets.

On the right side of Fig. , the pellets P2 do not densify to 70%. The trend of the curve shows, nonetheless, a similar behavior to the one registered in the same figure on the left side (as well as on the left side of Fig. 3).

Pellets P16-P24 were made in lab with the same raw material that was used for industrial production. In the manufacture of those pellets there was an assumed interest in assessing the behavior of the material pelleted at the factory, under pure compression. The sequence of figures allows noticing the overlapping effects of the characteristics of the materials. Thus, comparisons between P24 pellets made of broom and pine sawdust and their combinations, pellets P16 - P19, are shown in Fig. 6 to 9. The difference in the behavior of the same type of material but with different pre-treatment as can be seen in Fig.6 (left side), which shows the comparison curves of densification of pellets P1 and

P24 (10 mm diameter), broom with compression velocities of 1.5 MPa/s, pre-processed in the laboratory and factory. It can be observed that the lab pre-processed broom (P1) densified more than the industrially milled (P24). In the right side of the same figure, a comparison of the energy *vs* densification used for pellets P24 (red line) and P22 (green line) is presented. The difference of energy used by sawdust (P22) to a higher densification is apparent: half the energy used by broom (P24), which densified only 64%.

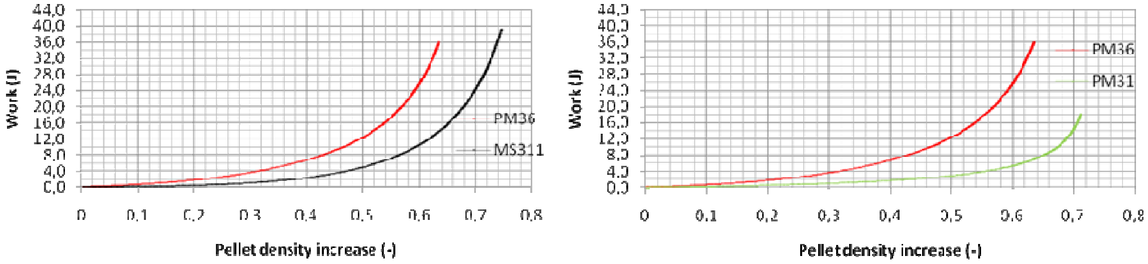

Figure 6. On the left side: densification curves of P1 and P24 pellets; on the right side: comparisons between the curves of densification and energy of the P24 (red line) and P22 (sawdust) pellets with 10 mm diameter and 1.5 MPa/s load rate.

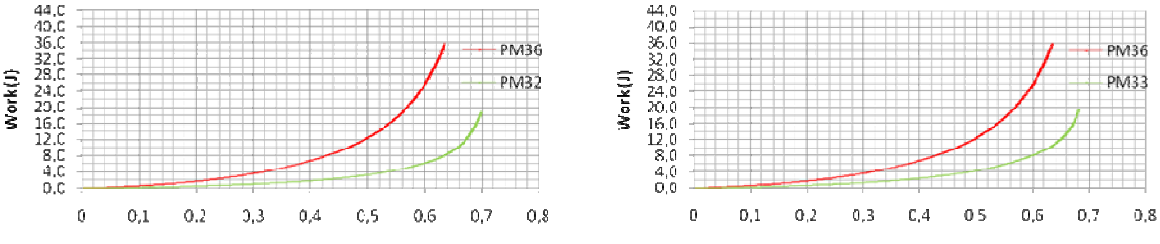

Figure 7. Energy spent on the formation of pellets with 10 mm diameter and 1.5 MPa/s load rate. Comparison between P24 (red line) and P16 (green line) pellets on left diagram; P24 (red line) and P17 (green line) pellets on right diagram.

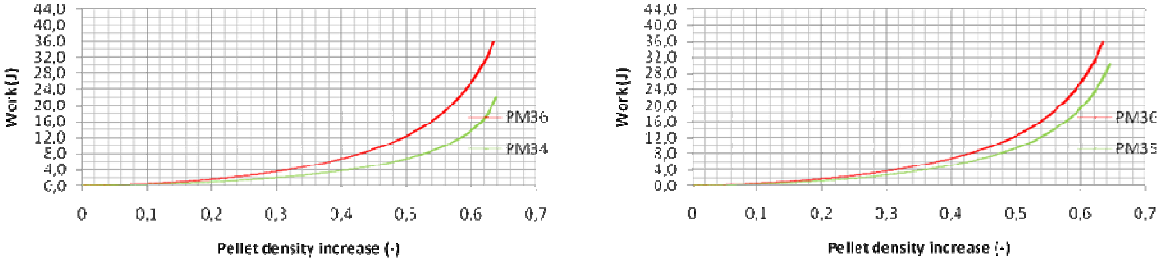

Figure 8. Energy spent on formation of pellets of 10 mm diameter 1.5 MPa/s load rate. Comparison between P24 (red line) and P18 (green line) pellets on left diagram; P24 (red line) and P19 (green line) pellets on right diagram.

It seems that the densification curve of the mixture of sawdust with broom moves upward as the broom percentage of the mixture increases, Fig. 17 to 20. It also seems that the behavior of the mixture subjected to pure compression approaches the predominant effect of the broom in the mix.

3.2. Pellets quality

The results of the characterization of some of the pellets produced are summarized in Tab.4:

Table 4. Characterization results of pellets.

Pellets	Diameter [mm]	Length [mm]	Unit mass [g]	Particle density [Kg/m^3]	Moisture content [wt %$_{ar}$ db]	Ash [wt %]	Fines [wt %$_{ar}$]	Bulk density [Kg/m^3]	Water absorbed [wt %]	Sample initial mass [g]	Sample final mass [g]	DU [wt %$_{ar}$]
P1	10.33	12.95	0.92	847.67	1.25	3.56	-	-	18.73	468	224.7	78.3
P 2	10.41	11.86	0.97	960.94	8.99	5.34	-	-	15.70	477	275.7	83.3
P 3	10.38	13.12	1.08	972.76	10.20	2.18	-	-	17.78	-	-	-
P 4	10.40	12.61	1.03	961.54	10.75	1.53	-	-	19.02	-	-	-
P 5	10.46	12.24	0.98	931.73	10.11	2.86	-	-	20.28	-	-	-
P 6	5.96	23.88	0.99	1486.00	43.48	3.76	1.6	663.0	16.16	554	539.4	99.1
P 7	5.84	23.28	0.97	1555.51	42.65	3.74	2.2	795.5	12.68	542	524.6	98.9
P 8	6.03	23.23	0.94	1416.95	36.23	1.33	2.0	763.0	12.18	589	574.2	99.2
P 9	5.99	22.74	0.69	1076.75	6.15	0.41	0.5	705.5	9.58	579	564.1	99.1
P 10	5.99	22.79	0.74	1152.24	4.23	0.97	0.9	775.5	11.45	542	526.9	99.1
P 11	5.94	26.35	0.81	1109.28	1.25	0.69	1.3	758.5	18.30	522	475.2	96.9
P 12	5.93	30.51	1.04	1234.22	1.96	3.29	1.8	735.0	16.71	546	534.1	99.3
P 13	6.02	32.30	0.97	1055.08	3.19	1.78	5.8	736.5	16.99	597	502.1	94.5
P14	6.01	31.08	0.96	1088.81	9.09	2.85	2.9	617.0	15.10	548	499.1	96.9
P 15	10.35	30.86	2.45	944.69	10.42	1.18	0.8	718.5	8.18	500	494.0	99.6

The European certification system "ENplus," classifies the properties of pellets in accordance with prEN 14961-2 on three levels, A1, A2 and B.

Table 5. Final classification of pellets industrially produced under de ENplus system.

Pellets	Dimensions class (D) [mm]	Moisture content (M) [w - % db]	Ash (A) [w - % db]	Fines (F) [w - %]	Bulk density (BD) [Kg/m^3]	Additives [w - %]	Net calorific value (Q) [MJ/kg]	[kWh/kg]	Mechanical durability (DU) [w - %]
P6	D06	43.48	A 5.0	F 2.0	BD 650	No	-		DU97.5
P7	D06	42.65	A 5.0	F 3.0	BD 795.5	No	-		DU97.5
P8	D06	36.23	A 1.5	F 2.0	BD 763.0	No	-		DU97.5
P9	D06	M10	A 0.5	F 1.0	BD 705.5	No	-		DU97.5
P10	D06	M10	A 1.0	F 1.0	BD 775.5	No	17.6	Q 4.9	DU97.5
P11	D06	M10	A 0.7	F 2.0	BD 758.5	No	18.2	Q 5.1	DU96.5
P12	D06	M10	A 5.0	F 2.0	BD 735.0	No	13.8	Q 3.8	DU97.5
P13	D06	M10	A 3.0	F 5.8	BD 736.5	No	-		DU94.5
P14	D06	M10	A 3.0	F 3.0	BD 600	No	-		DU96.5

The first items represents the highest level of quality, relevant to private end-users such stove owners. In class A2 the limits are less rigorous, allowing the use of pellets in units equipped with high capacity boilers. Class B does not allow the inclusion of treated wood, but admits pellets for industrial use with 'lower' properties than in Class A2.

Based on Tab. 4 data, an analysis of standardized prerequisites for the pellets produced was made. The results are presented in Tab.5. Since the raw material (rockrose and broom) used for the production of industrial pellets was milled with flowers and bark, and sawdust with left over industrial beech tape, possibly contaminated with minerals, the major hindrance of pellets made with broom and with rockrose is the excessive amount of ashes.

Table 5 indicates also that at least two types of pellets are liable to fit the requirements of Class A2 and B, pellets P10 and P11, respectively. Pellets P9 meet all the conditions to belong to the class A1 but, as its HHV was not determined, it was not classified.

4. CONCLUSIONS

Broom and rockrose are viable options to pellets production. The combinations of the two species with leftover wood industries and within the resulting standard limits may also contribute to obtain high quality pellets. A more comprehensive and deeper discussion of the results is envisaged in the near future. Namely, the stability in contact with moisture and the durability under conditions of mechanical wear are issues that will be revisited. Establishing a criterion that provides an assessment of the overall quality was not attained so far. Further work is needed and shall be developed under an approved R&D Government project (PTDC/AGR-CFL/114826/2009).

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