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Obtaining and characterization of pellets based on walnut shells, waste paper and paperboard

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Abstract

Spite of continuously deforestation in Chile in the last 40 years, the production of combustion pellets is increased, being the main producer Ecomas. For this study, it was tested walnut and wheat shells, waste paperboard and waste Kraft paper as raw materials for 17 formulations of pellets. Only 8 formulations were successful for pellet production. The 8 pellets obtained and Ecomas pellet were compared according to ash and moisture content and heating value (HHV). The pellets obtained indicated a better HHV and moisture content but a higher ash content than Ecomas pellet. Moreover, 8 pellets and Ecomas pellet were compared with the set values in European Norm EN 14961-2, indicating that pellets 5 and 6 meet the EN 14961-2. In view of the results achieved, the mixtures used in this study could considered as promising raw material for the production of combustion pellets with similar characteristics as commercial pellet Ecomas.

Keywords: Pellets, walnut shells, wheat shells, waste paperboard, EN 14961-2

Obtención y caracterización de Pellets basados en cascaras de nueces, residuos de cartón y papel

Resumen

A pesar de la continua deforestación en Chile en los últimos 40 años, la producción de pellets para la combustión se ha ido incrementando, siendo el principal productor Ecomas. Para este estudio, se propusieron 17 formulaciones basadasen cáscara de nuez y trigo, desecho de cartón y papel Kraft para la obtención de pellets. Sólo 8 formulaciones fueron exitosas para la producción de pellets, las cuales fueron comparadas con el pellet de Ecomas mediante el contenido de cenizas y humedad y el poder calorífico superior (HHV). Los pellets de este estudio indicaron un mejor contenido de HHV y humedad, pero un mayor contenido de cenizas. Además, los 8 pellets y el de Ecomas fueron comparados valores establecidos en la norma europea EN 14961-2, observando que el pellet de Ecomas, y el pellet 5 y 6 cumplen con EN 14961-2. En vista de los resultados obtenidos, las mezclas utilizadas (5 y 6) podrían considerarse como una potencial materia prima para la producción de pellets con características similares al pellet comercial Ecomas.

Palabras clave: Pellets, cáscaras de nuez, cáscaras de trigo, cartón residual, EN 14961-2
Introduction

There is a growing interest in substituting fossil fuels by renewable energies[1]. Pellets have received increasing attention not only for their potential as renewable energy but also to be considered a clean (reduction of particulate matter emissions by up to 50%) and affordable source [2-3]. Combustion pellets are a standardized solid biofuel of cylindrical shape, whose production is obtained by pressing the organic material. Their manufacturing provides an enhancement in the handling and storage and lower transport costs [4].

A recent study identified Chile as the most attractive Latin-American country for investments in pellet mills, offering the best results between availability of biomass, annual yields and low investment risk [5]. However, wood demand for pellets production could affect sustainability and management of forest by deforestation, detecting in Chile a continuously decrease of tree cover [6-7]. In this sense, investigation on alternative raw materials for pellets production is necessary to decrease pressure on natural resources.

Chilean government is becoming increasingly aware of the high level of air pollution caused by residential heating systems based on firewood, often of poor quality and high moisture content. Since 2016, the regional governments carried out a heating and wood policy which includes incentives to increase the regional use of combustion pellets, being the most commercialized pellet by Ecomas Company with 80% of national combustion pellets production [8-12]. Therefore, it is expected an increase on combustion pellets demand and consequently also on the pressure on wood.

This study focus on obtaining pellets composed by walnut and wheat shells, waste paperboard and Kraft paper with similar characteristics to commercial pellet from Ecomas. Resulting and Ecomas pellets will be characterized according to European norm EN 14961-2.

Materials and Methods

Materials

The raw materials for the present study were obtained in the Region “La Araucanía”, Chile (38°54′S 72°40′W). Two raw materials came from agro-industrial activities: walnut shells (WaS) and wheat shells (WsS), and the other two from solid household wastes: waste paperboard (WP) and waste Kraft paper (WKP).

Characterization of raw materials

Previous to the characterization, the four raw materials were milled (Retsch Mill, ZM200) at 6000 rpm equipped with a 500 µm sieve. The structural analysis consisted of a quantitative method based on gravimetric analysis to estimate extractives [TAPPI T-264 cm-07, [13]], cellulose (Jayme-Wise and Kushner and Hoffer method), hemicellulose and lignin (Klason method) [13-14]. The moisture (M) and ash content (A) was determined as the residue left after thermal treatment of raw materials at 105 °C and 575 ± 2.5 °C for 3.5 h in a muffle furnace, respectively. The quantification of higher heating value (HHV) was carried out using a Parr 6200 calorimetric bomb.

Formulations proposal and pelletizing

It was proposed 17 formulations with different doses of WaS, WsW, WP and WKP without a previous drying process. The formulation was based on the availability and HHV of raw materials (Figure 1). Moreover, sodium lignosulfonate between 4 and 10% (w/w) was added to enhance binding effect among the raw materials during pelletizing.

![Figure 1. Proposed formulations with the raw materials of this study and the sodium lignosulfonate (Na-L) as binder agent.](image_url)
Characterization of pellets

Among the formulations, those that visually met the desired dimensions were characterized and compared with Ecomas pellet. The characterization included: (i) moisture content, (ii) average length and diameter determined by measuring with caliper (randomly selected), (iii) bulk density determined by measuring the mass of a fixed volume filled with the pellets, (iv) ash content by 550 ± 2.5 °C for 3.5 h in a muffle furnace, (v) the compressibility strength of the pellets by the determination of the force at break (Compression Tester Egeo) and (vi) the determination of HHV using Parr 6200 calorimetric bomb [15-16].

Results and Discussion

Characterization of the raw materials

The quality of the pellets for thermochemical conversion utilization depends on the physical-chemical (Table 1 and 2) of the raw materials. Therefore, inquiry about chemical composition of raw materials from the present study, would be information about their potential to form pellets.

Table 1. Moisture content and HHV for raw materials used in this study.

| Samples | M* (%) | HHV* (MJ/kg) | References consulted |
|---------|--------|--------------|---------------------|
| WaS     | 8.1 ± 0.7 | 19.6 ± 1.1  |                     |
| WsS     | 10.9 ± 0.2 | 18.3 ± 1.0  |                     |
| WP      | 5.4 ± 0.7  | 17.8 ± 1.3  | [17-18]             |
| WKP     | 5.0 ± 1.4  | 16.6 ± 1.1  |                     |

*M: Moisture, HHV: Higher heating value

Table 1 provides the results of moisture content and HHV. Both properties were compared with data reported in scientific literature, indicating that almost all values are in the range reported for such raw materials. Among the raw materials, WsS presented the higher value of moisture content which could be attributed to the presence of hemicelullosse, because the presence of this structure promotes water absorption. For pelleting it is preferred a low content of water to improve the resistance of the pellets to crumbling effect. Nevertheless, the presence of water during pelleting is necessary for the development of intermolecular forces between the particles from raw materials. In addition, the HHV is also affected by a high moisture content. The higher the value of moisture, the lower the amount of energy produced and therefore the lower the quality of the resulting pellet.

However, according to the literature reviewed, the effect of moisture on densification is critical but is not well understood, acting water as binder and also as lubricant [19].

Due to almost all values of moisture content were below 10%, it was decided to desist from the drying pre-treatment before pelleting process. Moreover, it is expected that the high temperature in pelleting machine, and the friction between the mixture of raw materials and the complex of die and cylinder could cause a significant reduction of the moisture content of resulting pellets by evaporation.

Regarding HHV, all raw materials showed values higher than 16.6 MJ/kg. Raw material WaS stands out due to the high HHV. Authors reported that there are a positive and significant correlation between the lignin content and the HHV [20]. As observed in Table 2, WaS presented also the higher value of lignin content.

The chemical composition based on the structural composition of raw materials is illustrated in Table 2, indicating that obtained values are within the values reported in literature [17]. The percentage of extractables reached higher values for WP and WKP, exceeding 13%, whereas WaS and WsS presented lower values than 11%. The presence of extractables in the raw materials such as waxes and resins could influence the pelleting and the pellet quality, favoring the crumbling effect [19]. As expected, recycling raw materials WP and WKP show a lack of lignin and hemicellulose and high values of cellulose, due to these materials come from the cellulose production, after delignification process.

Table 2. Chemical composition of raw materials and ash content for all raw materials tested, expressed in percentage (% w/w).

| Samples | Extractables | Hemicellulose | Cellulose | Lignin | Ash |
|---------|--------------|---------------|-----------|--------|-----|
| WaS     | 11.7         | 5.4           | 27.0      | 46.5   | 1.2 |
| WsS     | 9.2          | 46.5          | 8.6       | 21.7   | 3.3 |
| WP      | 14.2         | 0             | 75.8      | 0      | 4.8 |
| WKP     | 13.5         | 0             | 69.8      | 0      | 11.8|

*References consulted for range reported: [17-18, 21-22]

The percentage of hemicellulose was higher for WsS, which corroborates relationship between water and hemicellulose content [23]. The amount of lignin reached the highest value for WaS. This result could contribute to a better structural stability of the resulting pellets. In case of the ash content, almost raw materials evidenced a low value. The exception was WKP with a value of 11.8%, which could imply a high ash content for the resulting pellets.
pellets based on certain quantities of WKP.

**Obtaining of pellets from proposed formulations**

The current study found that from all formulations tested (Figure 1), only 8 formulations were possible for pellet production (Formulations 1 to 8). The formulations without addition of the binder agent (Na-L) and formulations based 100% of WaS, WsS, WP and WKP failed. Moreover, pellets (Formulations 9 to 17), observing a strong crumbling effect in the resulting pellets.

Figure 2. Photos of the obtained pellets with the formulations 1 to 8.

Figure 2 shows the photos of the formulations with a successful pelleting process. These 8 formulations are composed by all raw materials. This result indicate that the contribution of lignocellulosic materials, cellulosic materials and small addition of Na-L, favors the bonding of particles by intermolecular attractive forces (e.g. hydrogen bonds).

The characterization of the pellets are illustrated in Table 3. All pellets revealed adequate levels of moisture content, heating value and bulk density (> 550 kg/m³). Pellets obtained presented similar diameter and lengths to Ecomas pellets. Regarding moisture content, pellets showed values (< 2%) lower than Ecomas pellet.

It was observed that the higher the mass contribution of WaS and WsS in the formulation, the higher the water content in the resulting pellets, which could be attributed to the high water content of both raw materials. The pellets obtained confirm that the amount of lignin of the raw materials directly influences the quality of the pellet, providing an increase in HHV.

From the results, it also detected that the ash content in the pellets tends to increase with an increasing of the Na-L dose. Moreover, the ash content values in the pellets are higher than the value of Ecomas pellet, but lower than the value set in the standard norm.

The pellets obtained confirm that the lignin content in the raw materials influences directly the quality of the pellet, providing an increase in HHV. In addition, an increasing of WaS and Na-L dose improve considerably the HHV for the pellets obtained. This statement corroborate study carried out by Demirbas [20]. It is clear, that both values are of great concern for the combustion process in domestic stoves. From these results, could be suggested that combustion would generate low particulate matter emissions.

Regarding the compressibility strength, the values were sufficiently high to remain intact and within values reported by Puig-Arnavat et al. [19]. This parameter behaves with similar trend as the ash content, showing values between 3.5 and 9.1 kN.

In order to enable a better visualization (Figure 3), the values of HHV, moisture, bulk density and ash content of all pellets were compared with the set values for the standard norm EN 14961-2[24].

Among the pellets obtained, the moisture con-
Table 3. Results from the characterization of the pellets obtained from formulations 1 to 8.

| Pellets | Diameter (mm) | Length (mm) | M* (%) | HHV* (MJ/kg) | A* (%) | BD* (kg/m³) | Compressibility Strength (kN) |
|---------|---------------|-------------|--------|--------------|--------|-------------|-------------------------------|
| 1       | 6 ±0.1        | 21 ±0.3     | 0.04 ±0.03 | 20.2         | 7.0 ±0.19 | 556 ±0.02 | 5.2 ±0.02                      |
| 2       | 6 ±0.2        | 21 ±0.2     | 1.88 ±0.05 | 17.4         | 5.0 ±0.23 | 590 ±0.02 | 6.2 ±0.01                      |
| 3       | 6 ±0.3        | 23 ±0.3     | 0.27 ±0.04 | 20.4         | 7.9 ±0.05 | 586 ±0.02 | 6.21 ±0.01                     |
| 4       | 6 ±0.1        | 23 ±0.1     | 0.75 ±0.04 | 18.2         | 5.2 ±0.17 | 579 ±0.02 | 6.0 ±0.01                      |
| 5       | 6 ±0.1        | 27 ±0.2     | 0.29 ±0.01 | 21.3         | 6.7 ±0.18 | 619 ±0.02 | 9.1 ±0.02                      |
| 6       | 6 ±0.2        | 25 ±0.1     | 0.41 ±0.02 | 18.5         | 5.5 ±0.21 | 607 ±0.02 | 6.2 ±0.01                      |
| 7       | 6 ±0.2        | 21 ±0.1     | 0.45 ±0.02 | 18.8         | 6.2 ±0.13 | 586 ±0.02 | 6.1 ±0.01                      |
| 8       | 6 ±0.2        | 17 ±0.1     | 0.55 ±0.02 | 18.4         | 4.3 ±0.22 | 584 ±0.02 | 3.5 ±0.02                      |
| Ecomas  | 6 ±0.09       | 25 ±0.1     | 6 ±0.01   | 17.9         | 0.2 ±0.15 | 640 ±0.2 | 11.2 ±0.01                     |

*M: moisture content; HHV: higher heating value; A: ash content

tent of all pellets were below 15% and 6%, corresponding to EN 14961-2 and Ecomas pellet. In case of ash content, all pellets obtained meet the EN 14961-2, whereas regarding bulk density only pellet 5 and 6 presented similar values as the set value of EN 14961-2. Regarding HHV, only pellet 2 was below the set value of EN 14961-2 (18 MJ/kg). The other pellets showed an increase of 11% and 16% compared with Ecomas pellet and the EN 14961-2[24]. These preliminary results (Table 3 and Figure 3) indicates that pellet 5 and 6 are suitable and could compete in the regional pellet market with Ecomas pellet. Moreover, both pellets meet EN 14961-2[24]. Therefore, the formulations could be recommended for the pellets production.

However, the authors are convinced that the results obtained in this research are the first step for the commercial use of pellet 5 and 6. Further experimental investigations are needed to improve the pelletizing process.

Conclusions

The current findings in this study are relevant for the development of the pellet market in the Chilean region “La Araucanía”. The evidence suggests that walnut shells, wheat shells, waste paperboard and waste Kraft paper are potential raw materials for the production of pellets. In addition, the obtaining of pellets based on these raw materials could present a high potential to improve air quality. The most relevant finding of this study emerge directly from the results, pellets 5 and 6 meet EN 14961-2 and present higher values of the parameter tested compared with the current commercial pellet Ecomas.

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