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New thermal insulation fiberboards from cake generated during biorefinery of sunflower whole plant in a twin-screw extruder



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ABSTRACT

The objective of this study was to manufacture new thermal insulation fiberboards by thermo-pressing. The starting material was a slightly deoiled cake (17.6% oil content), generated during the biorefinery of sunflower (Helianthus annuus L.) whole plant in a co-rotating (Clextral BC 45, France) twin-screw extruder. All fiberboards produced were cohesive mixtures of proteins and lignocellulosic fibers, acting respectively as binder and reinforcing fillers in what could be considered as a natural composite. The molding experiments were conducted using a 400 ton capacity heated hydraulic press (Pinette Emidecau Industries, France). The influence of molding conditions on board density, mechanical properties and heat insulation properties was examined. Molding conditions included mold temperature (140-200°C), pressure applied (150-250 kgf/cm²) and molding time (40-76 s), and these greatly affected board density and thus the mechanical and heat insulation properties. Board density increased with increasingly extreme molding conditions, rising from 500 to 858 kg/m³. The mechanical properties increased at the same time (from 52 to 660 kPa for flexural strength at break, from 5.9 to 49.4 MPa for elastic modulus, from 0.5 to 7.7 kJ/m² for Charpy impact strength, and from 19.2 to 47.1° for Shore D surface hardness). Conversely, heat insulation properties improved with decreasing board density, and the lowest thermal conductivity (88.5 mW/mK at 25 °C) was obtained with the least dense fiberboard. The latter was produced with a 140 °C mold temperature, a 150 kgf/cm² pressure applied and a 40 s molding time. A medium mold temperature (160 °C) was needed to obtain a good compromise between mechanical properties (272 kPa for flexural strength at break, 26.3 MPa for elastic modulus, 3.2 kJ/m² for Charpy impact strength, and 37.3° for Shore D surface hardness), and heat insulation properties (99.5 mW/mK for thermal conductivity). The corresponding board density was medium (687 kg/m³). Because of their promising heat insulation properties, these new fiberboards could be positioned on walls and ceilings for thermal insulation of buildings. The bulk cake also revealed very low thermal conductivity properties (only 65.6 mW/mK at 25 °C) due to its very low bulk density (204 kg/m³). It could be used as loose fill in the attics of houses.

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1. Introduction

Sunflower (Helianthus annuus L.) is cultivated for the high oil content of its seeds. Oil represents up to 80% of its economic value. The industrial process for oil production consists of four successive stages: trituration, pressing, extraction of the residual oil using hexane, and refining (Isobe et al., 1992; Rosenthal et al., 1996). Extraction yields are close to 100% with very good oil quality. However, the use of hexane for oil production is an increasingly controversial issue and could be prohibited due to its

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carcinogenicity (Galvin, 1997). Consequently, numerous solvents have been considered, including water (Rosenthal et al., 1996).

Several researchers have studied the aqueous extraction of sunflower oil (Evon et al., 2007, 2009; Hagenmaier, 1974; Southwell and Harris, 1992) that is an environmentally friendly alternative to solvent extraction. It can be conducted using whole seeds (Evon et al., 2007) or from a press cake (Evon et al., 2009) in a Clextral BC 45 (France) co-penetrating and co-rotating twin-screw extruder that enables efficient mechanical lysis of the cells. Three essential unit operations are carried out in a single step and in continuous mode: conditioning and grinding of the initial material, liquid/solid extraction, and liquid/solid separation. A filter section is positioned on the barrel to collect an extract (filtrate) and a raffinate (cake), separately. However, the introduction of a lignocellulosic residue upstream from the filtration module is essential to enable liquid/solid separation.

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When applied to the whole plant, aqueous fractionation in a twin-screw extruder does not require the addition of a lignocellulosic residue (Evon et al., 2010a) due to the natural abundance of fibers in sunflower stalk (Maréchal and Rigal, 1999), and twinscrew extrusion technology thus appears to be an original and powerful solution for the biorefinery of sunflower whole plant. Under optimal operating conditions, oil extraction yield reaches 57%, and residual oil content in cake is 14.3% of the dry matter. These conditions lead to the co-extraction of proteins but also pectins and hemicelluloses. The corresponding protein extraction yield is 44%, and residual protein content in cake is 7.3% of the dry matter.

Cake moisture content is relatively high (at least 62%), and so it is first dried to facilitate conservation. It has a porous structure, and is mainly composed of lignocellulosic fibers (around 58% of the dry matter), but also cell debris from the kernel breakdown process. Actually, it is lixiviated matter where soluble molecules (proteins, pectins. . .) and lipids are partly removed, although plant structural molecules are not extracted. The cake is thus suitable for use in animal feeds and for energy production in pellet burning furnaces. Nevertheless, new valorizations of the cake as a mixture of proteins and lignocellulosic fibers can be also considered (Orliac et al., 2002, 2003; Rouilly et al., 2001, 2003, 2006a,b).

The cake's thermo-mechanical behavior has been previously studied (Evon et al., 2010b). DSC (differential scanning calorimetry) measurements indicate that denaturation of cake proteins is almost complete, and the DMTA (dynamic mechanical thermal analysis) spectrum obtained from dried and ground samples reveals a significant peak at high temperature (around 180 °C) which, as already observed with industrial sunflower cake (Geneau, 2006), is associated with the glass transition of proteins. Because the cake is a mixture of proteins and lignocellulosic fibers, it can be considered as a natural composite, and can thus be successfully processed into cohesive, biodegradable and value-added fiberboards by thermopressing, proteins and lignocellulosic fibers acting respectively as internal binder and reinforcing fillers (Evon et al., 2010b, 2012a).

The mechanical properties for bending of fiberboards, increase with temperature, pressure and length of time of thermo-pressing (Evon et al., 2010b, 2012a). The highest flexural strength at break (11.5 MPa) and the highest elastic modulus (2.2 GPa) are obtained from a cake with a dry matter residual oil content of 14.5%, and under the following molding conditions: 500 mg/cm^2 for cake quantity, 200 °C for the temperature of the two aluminium plates of the heated hydraulic press, 320 kgf/cm² for pressure applied, and 60 s for molding time (Evon et al., 2010b). The fiberboard thickness is only 3.9 mm, its density is quite high (1035 kg/m^3) and DMTA analysis reveals a significant vibratory oscillation peak at low temperature (between -20 and -14 °C), which is attributed to the β -transition of proteins (glass transition of their side chains) (Rouilly et al., 2006b; Zhang et al., 2001). No significant transition is observed between 0 °C and 200 °C, meaning that proteins ensure the agromaterial's cohesion without any phase change in this temperature range. Finally, lignocellulosic fibers' entanglement also act as reinforcement. Because of its promising flexural properties, such a fiberboard would be potentially usable as inter-layer sheet for pallets in the handling and storage industry or for the manufacture of biodegradable, multi-board containers, e.g. composters, crates for vegetable gardening (Evon et al., 2010b).

Another industrial application of fiberboards made from renewable resources is heat insulation of buildings (walls and ceilings), where the main advantages of vegetable fibers are abundance, low cost (the majority are agricultural residues), minimal environmental impact, independence from fossil resources, and their natural capacity for thermal insulation (Saiah et al., 2010). Insulation boards can be made from maize husks and cobs (Paiva et al., 2012; Pinto et al., 2011; Sampathrajan et al., 1992), a mixture of durian peels and coconut coir fibers (Khedari et al., 2003, 2004), cellulose (Nicolajsen, 2005), wastes from tissue paper manufacturing and corn peel (Lertsutthiwong et al., 2008), kenaf fibers (Ardente et al., 2008), flax and hemp fibers (Benfratello et al., 2013; Korjenic et al., 2011; Kymäläinen and Sjöberg, 2008), cotton stalk fibers (Zhou et al., 2010), jute fibers (Korjenic et al., 2011), coconut fibers (Alavez-Ramirez et al., 2012; Panyakaew and Fotios, 2011), sunflower pith (Vandenbossche et al., 2012), date palm fibers (Chikhi et al., 2013), etc.

The thermal conductivity of insulation boards is often influenced by their densities (Benfratello et al., 2013; Chikhi et al., 2013; Khedari et al., 2003, 2004; Lertsutthiwong et al., 2008; Panyakaew and Fotios, 2011; Vandenbossche et al., 2012; Zhou et al., 2010), and low-density materials have the lowest thermal conductivities. As an example, the thermal conductivity of an insulation board from sunflower pith is only 38.5 mW/mK at 25 °C with a board density of 36 kg/m³ (Vandenbossche et al., 2012). It is comparable to that of conventional insulation materials like expanded polystyrene $(37.4 \text{ mW/mK} \text{ with a board density of } 50 \text{ kg/m}^3)$, rock wool (35.6 mW/mK with a board density of 115 kg/m³), and glass wool (35.4 mW/mK with a board density of 26 kg/m^3). Thermal conductivity is higher with medium-density materials: 46-68 mW/mK at room temperature for coconut husk insulation boards with board densities of 250–350 kg/m³ (Panyakaew and Fotios, 2011), 81.5 mW/mK for a cotton stalk fibers insulation board with a board density of 450 kg/m^3 (Zhou et al., 2010), 89.9-107.9 mW/mK for hemp fibers insulation boards with board densities of 369–475 kg/m³ (Benfratello et al., 2013), 103.6 mW/mK for a coconut coir insulation board with a board density of 540 kg/m³ (Khedari et al., 2003), and 150 mW/mK for a date palm fibers insulation board with a board density of 754 kg/m^3 (Chikhi et al., 2013). Nevertheless, such boards are viable options for use in building insulation (walls and ceilings).

Heat insulation properties, of fiberboards from cake generated during the biorefinery of sunflower whole plant in a twin-screw extruder are also promising, even if the corresponding board densities are quite high (904–966 kg/m³) (Evon et al., 2012b). Indeed, thermal conductivity at 25 °C is rather low for the three fiberboards tested, and it decreases from 135.7 to 103.5 mW/m K with an increase in board thickness from 5.4 to 10.2 mm. Thus, the thickest fiberboard (density 917 kg/m³) gives the best thermal insulation. Fiberboards from such a cake with lower density and higher thickness would perhaps produce a significant improvement in their heat insulation properties.

This study aimed to manufacture by thermo-pressing, new thermal insulation fiberboards with medium density (from 500 to 900 kg/m^3), and high thickness (more than 10 mm and up to 20 mm), from cake generated during the biorefinery of sunflower whole plant in a twin-screw extruder, and to evaluate the influence of molding conditions (temperature, pressure, and time) on their mechanical (flexural properties, Charpy impact strength, and Shore D surface hardness) and heat insulation properties.

2. Materials and methods

2.1. Material

Thermo-mechanical fractionation in the twin-screw extruder was carried out using a batch of sunflower (*Helianthus annuus* L.) whole plant of the oleic type (La Toulousaine de Céréales, France) (Table 1), harvested in September, i.e. at plant maturity. Whole plant was previously dried in a ventilated oven $(50 \degree C, 48 h)$ and crushed using a hammer mill (Electra VS 1, France) fitted with a 15 mm screen. The moisture content of the powdered plant (batch of around 250 kg) was $7.2 \pm 0.1\%$ (French standard NF V 03-903).

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