



# Characterization and application of a natural polymer obtained from *Hydrangea macrophylla* as a thermal insulation biomaterial



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## ABSTRACT

Thermal insulation materials play a significant role in improving the energy efficiency of buildings and therefore many studies focus their efforts on developing low environmental impact materials. In this context, the aim of this work was to study a natural polymer obtained from *Hydrangea Macrophylla* (HM) and its application as a thermal insulation material. The natural polymer was chemically, physically and mechanically characterized and compared to polyurethane (PU) and expanded polystyrene (EPS). In addition, a composite material blended in a mold using HM stems and sprayed with PU was used to evaluate possible applications. The results obtained from morphological studies conducted on HM showed it to be an interesting natural porous structure with a smooth surface similar to those found in closed cell insulation materials and highly compatible with PU foam. This composite showed good thermal stability, an outstanding thermal conductivity for a natural insulation material, high density and good behavior under compression. HM polymer in a PU matrix like a block type was shown to be an appropriate thermal insulation material, providing an excellent opportunity to reduce the environmental impacts of these types of insulating foams.

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## 1. Introduction

Buildings are the largest consumers of energy and the most significant contributors to climate change greenhouse gas emissions in the United States [1]. In addition, the stages of construction and use of buildings in the European Union (EU) accounts for about half of all their extracted materials and energy consumption and about a third of their water consumption and all waste generation [2]. Meanwhile, in Chile, the energy consumption of the public, commercial and residential sectors represents a 26% of the country's final energy consumption [3]. For all these reasons, this sector is associated with environmental pressures that arise at different

stages of a building's life-cycle, including the manufacturing of construction products, building construction, use, renovation and the management of building waste. Buildings consume energy directly or indirectly in all stages of their life cycle right from the cradle to the grave, and there is an interplay between the stages of energy use (embodied and operating energy). Hence, they need to be analyzed from a life cycle point of view. Some case analysis found in the literature show that life cycle energy use of buildings depends on the operating (80–90%) and embodied (10–20%) energy of the buildings [4,5]. A buildings life cycle energy demand can be reduced by decreasing its operating energy using high performance insulation materials. However, such a reduction in operating energy demand is usually accompanied by increase in embodied energy of the building due to intensive energy consumption of insulation materials during the production process [4]. As insulation materials play a significant role improving energy efficiency in buildings, many studies have focused their efforts on developing low environmental impact materials. These include, natural fibers obtained from plant stems, leaves, roots, fruits and seeds of plants

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[6] or polymeric materials such as lignocellulosic materials, which have numerous advantages and are the most promising in terms of environmentally friendly and renewable materials in the building sector [7–10]. Agricultural waste is produced in large quantities worldwide, and could therefore be one of the most interesting renewable materials for use in thermal insulation [11], reducing the operational energy, and environmental impacts in their manufacture, and final disposition [12]. Several researchers have studied the thermal conductivity of different plant fibers composites, including fibers arrangement and surface treatment [13]. Thus, agricultural fibrous materials such as straw, flax, cotton and hemp have been investigated as possible insulation products, and all are examples of the trend towards sustainable materials in building construction. No doubt, natural fibers, such as hemp, flax, cotton, jute, sisal, pineapple and cereal straw, can be used in a variety of manners. In this context, natural fibers or polymers have a high potential to be used in bio-based materials for different applications [14–17]. An interesting natural porous structure is present in the *Hydrangea macrophylla* plant. It is a rounded deciduous shrub and native of northeast Asia, which is cultivated in many parts of the world and in various climates. It is a popular ornamental plant in Europe, and about 17 million potted plants are produced annually. Germany is the main producer (39%), with France in second place (27%) followed by the Netherlands, Italy and Spain [18]. In Chile, it is a very abundant and also a very popular ornamental plant in cities and rural areas. Meanwhile, exports in 2011 accounted for 2.36% of all exported cut flowers [19]. Considering his fast growing, abundant, and potential farming to produce ornamental flowers and use the stem of the prune, the aim of this work was to characterize a natural polymer obtained from *Hydrangea macrophyll* and its application as a possible thermal insulation material.

## 2. Materials and methods

### 2.1. Materials

The natural polymer characterized in this study was obtained from the plant *Hydrangea macrophyll* (HM), var. macrophylla. This plant was sampled in Huichahue, a rural locality near Temuco city in the Region of the Araucana in southern Chile. Stems from HM plants were collected manually and air dried for two weeks. The stems were then peeled and finally stored for further analysis (Fig. 1). These stems were on average of  $40.2 \pm 21.3$  mm in length and  $6.0 \pm 1.7$  mm in diameter. A commercial polyurethane (PU) with isobutane and propane gas used as the blowing agent, and a commercial expanded polystyrene (EPS) with pentane gas as the blowing agent, were used for comparison purposes with commercial polymers used as traditional thermal insulation materials in buildings envelopes. In addition, a composite material blended in a mold using HM stems and sprayed with PU was used to evaluate possible applications. A molding process was used to form the blocks of PU and HM + PU. A mold with a lid as shown in Fig. 2-A was used. The process can be described in three steps; 1) Stems were added in a mold with a polyethylene layer for easy demolding; 2) PU spray was added among the holes generated by the stems; 3) A lid with a weight was used above to prevent the stems from rising. The result was a block as shown in Fig. 2-B; then samples were cut as can be observed in Fig. 2-C in the case of a flexural sample.

### 2.2. Chemical composition

Chemical analysis of the fibers was performed according to TAPPI procedures and other experimental procedures described by Wisse et al. [20] and Rowell [21]. The sample preparation for

further analysis was done following the procedure TAPPI T 257 cm-85 procedure; water content determined by using Tappi T264 cm-97; ashes content following TAPPI T211 om-93 and lignin by TAPPI method T222 om-98. The holocellulose content of the samples was determined according to the procedure described by Wise et al. [20]. Briefly, to  $2.5 \pm 0.1$  g of samples extractive (determined using TAPPI T204 cm-97) and moisture free samples were added 80 mL of hot water at ( $70 - 80^\circ\text{C}$ ) and heated in a water bath at  $70^\circ\text{C}$ , shaking periodically. After 60 min, for a period of 6 – 8 h, 0.5 mL of acetic acid and 2.6 mL of sodium chlorite (25%) were added. After the additions, the sample was maintained for 12 further hours without any addition. Finally, the residue was filtered and dried at  $105^\circ\text{C}$  and weighed. Cellulose and hemicellulose were determined following the method described by Rowell [21],  $2.0 \pm 0.1$  g of sample obtained from the holocellulose determination was mixed with 10 mL of NaOH (17.5%) by shaking until dispersed particles appeared. Afterward, there were three more additions of 5 mL of NaOH (17.5%) in a period of 15 min; the sample was maintained for 1 h and finally filtrated. Cellulose was calculated as a percentage of sample before treatment with NaOH and hemicellulose as the difference between holocellulose and cellulose.

### 2.3. Density

Densities of raw and processed HM samples were determined according to a modified process based on the ASTM D1622-14 standard. HM raw material density was obtained by weighing pieces of  $0.86 \pm 0.06$  cm<sup>3</sup>. Meanwhile, HM + PU, PU, and EPS densities were obtained using cubic pieces of 1 cm<sup>3</sup>. The values presented correspond to the average density determined for eight specimens of each sample measured as bulk raw material, longitudinally or transversely.

### 2.4. Thermal stability

The thermal stability of each sample was determined by measuring the global mass loss at different temperatures with a thermogravimetric analyzer (TGA) model STA 6000 from Perkin Elmer Co. Measurements were performed under nitrogen atmosphere. Temperature changes were controlled in the range between 20 and  $600^\circ\text{C}$  at a heating rate of  $10^\circ\text{C}/\text{min}$  for 60 min. The degradation temperature and maximum degradation rate were also calculated.

### 2.5. Surface analysis

A variable pressure scanning electron microscope (VP-SEM) Hitachi SU 3500, was used to compare the microstructure of the HM polymer and commercial insulation materials. Samples were observed in cross section. A backscattered electron detector (BSE) in compositional mode was used to analyze the surface of each sample under operating conditions of 3 kV and 15 Pa.

### 2.6. Infrared spectroscopy analysis

Fourier transform infrared spectroscopy (FTIR) analysis of each sample was performed. For this analysis, samples were loaded into the sample holder of a Cary 630 FTIR spectrometer (Agilent Technologies). Resolution of the spectrophotometer was set at  $4\text{ cm}^{-1}$ , and 140 scans were obtained in the  $4000 - 600\text{ cm}^{-1}$  spectral region.

### 2.7. Thermal conductivity

The thermal conductivity of each material was determined by

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