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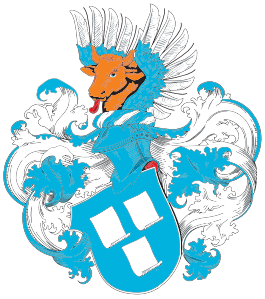

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VALORISATION OF BRAZILIAN PINE-FRUIT SHELL RESIDUE (*ARAUCARIA ANGUSTIFOLIA*) IN THE PRODUCTION OF BIOPOLYOLS

Stephany C. de Rezende[1,2], João A. Pinto[1], Fernanda V. Leimann[2], Maria F. Barreiro[1*]

[1] LSRE-LCM Associate Laboratory, Polytechnic Institute of Bragança, Campus Santa Apolónia Ap. 1134, 5301-857 Bragança, Portugal, *barreiro@ipb.pt, Phone/Fax: +351 273303089/+351 273313051
[2] Programa de Pós-Graduação em Tecnologia de Alimentos (PPGTA), Universidade Tecnológica Federal do Paraná – UTFPR Campus Campo Mourão, Via Rosalina Maria Dos Santos, 1233. CEP 87301-899. Caixa Postal: 271, Campo Mourão - PR - Brasil

ABSTRACT

The valorisation of agro-forestry residues, namely the ones which do not have commercial value, have attracted the interest of academia and industrials contributing to generate new alternatives in the field of chemicals and materials. The purpose is not only promoting their reuse to generate value-added products, but also highlight the production of bio-based products from renewable resources. *Araucaria angustifolia* is a coniferous tree native of South America, found in the South and Southwest of Brazil, part of a whole vegetation of the Atlantic Forest. The Brazilian pine-fruit shell (PFS) is an attractive lignocellulosic residue with potential to be used as a raw material to produce polymeric materials. In this context, this work comprises the characterization of PFS and their use to produce polyols trough oxypropylation.

The oxypropylation reaction was carried out in bulk in a pressure reactor using a set-point temper ture of 160 ºC. Three series were chosen (PFS/propylene oxide (PO) (w/v, g/ml) of 30/70, 20/80 and 10/90) using four levels of catalyst content (5, 10, 15 and 20%, (w/w, PFS based)). Maximum pre sure and temperature, and total reaction time (time to achieve a relative pressure of zero) were registered. The obtained polyols were characterized in what concerns unreacted biomass (UR), homopolymer content (content of PO oligomers resulting from the secondary homopolymerization rea tion), hydroxyl number (IOH) and viscosity (μ, 20°C).

Oxypropylation occurred at moderate conditions of temperature, pressure and time giving rise to liquid polyols with a homopolymer content ranging from 4-65%, a hydroxyl number between 257-605 mg KOH/g and very high viscosities for the series 30/70 (>500 Pa.s, 20°C). For the series 10/90 a maximum in homopolymer content was achieved (64.5%). For the series 30/70 a slight decrease of the homopolymer content was observed as the catalyst content increases. In a general way, IOH increased with the increase of catalyst content, apart from the series 20/80 where the IOH reach the maximum with 10% of catalyst (605.4 mgKOH/g). For the same catalyst content, viscosity increases with the increase of PFS/PO ratio.

The unreacted PFS residues varied between 3.6 and 77.4% of the original used biomass. The series 30/70 was the one that presented the higher amount of unreacted PFS (34.9-77.4 %) independently of the used catalyst amount.

In a general way it was observed that low PFS/PO ratios (i.e. 10/90) give rise to polyols with low unreacted PFS residue, low viscosity (due to a high homopolymer content), and the lowest IOH number (256.6 mgKOH/g). The polyols based on the PFS/PO ratio of 30/70 presented high unreacted PFS residues (34.9-77.4% (w/w)), low homopolymer content (4.4-12.1% (w/w)), high IOH number (the highest value correspond to the formulation 30/70/20, 605.4 mg KOH/g) and high viscosity (>500 Pa.s, 20°C).

To the best of our knowledge no other works concerning Brazilian pine-fruit shell oxypropylation are available in the literature. In this context, the results presented here pointed out for the viability of using this agro-forestry residue to produce bio-based polyols.

Keywords: Agro-forestry residues, Brazilian pine-fruit shell (PFS), oxypropylation, polyols.