

# Revitalizing lignin

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## Impact paragraph

The transition from a linear economy to a circular economy is recognized as a crucial way to achieve the 2050 climate neutrality targets of the Paris Agreement.<sup>1, 2</sup> As a result of shifting toward circular economy, the use of biobased starting materials is gaining more and more interest from both industry and academia.<sup>3-6</sup> Here, I intend to highlight the impact of this research on academia, industry, and society.

The ultimate goal of this work was to valorize and benchmark lignin fractions in various applications, including adhesives, coatings, additives, and adsorbents. This research utilized lignin fractions originating from different biomass feedstocks, obtained via different processing methods such as state-of-the-art isolation of technical lignins, solvolysis of technical lignin, 'lignin-first' process, and separation of lignin oils. It is worth to highlight that those fractions were obtained as a result of cross-border collaboration within BIO-HArT and InSciTe Lignin RICHES platforms. Those lignin isolation technologies are currently upscaled or will be upscaled in the near future.<sup>7-9</sup> It is crucial to be pointed out that most of the applications for lignin-based materials are currently developed using technical lignins, possessing higher molecular weights, lower purity, and lower functionality. Therefore, the above-mentioned lignin fractions are significantly different from the commercially available technical lignins, as demonstrated in this thesis. Consequently, valorization of the above-mentioned lignin fractions in different applications is relevant for their commercialization and development of their marketable applications. In this work, I have shown some examples of potential applications in the field of polymeric materials and additives. Having said that, this work is relevant for both academic research, as evaluation of novel lignin fractions, and for industry, by providing a possible application pool of the lignin fractions obtained by modern biorefineries. In the future, the applications of the above-mentioned lignin fractions should not be limited to the ones presented here, but can be further developed and include more types of polymeric resins, such as polyurethane and epoxies. In addition, different applications could be tested, for example, polymeric blends, alternative purpose additives, and surfactants.

Another aspect of the research presented in this thesis is related to the development of suitable methodologies for incorporation of lignin fractions in polymeric materials and additives. Although there are already numerous existing methodologies, this work presents alternative approaches utilizing thiol-yne 'click' chemistry and optionally post-curing for the synthesis of lignin-based resins and lignin esterification for the biolubricants additives. The work demonstrated in this thesis shows how the properties of such materials can be tuned and how those materials compare with state-of-the-art references. Even though the methodologies presented in this thesis

were shown to be suitable for modifying (biobased) materials, their application combined with real-life applications was not demonstrated for lignin-based materials. In my opinion, especially valuable are the results shown in chapter 3. The most surprising result of that chapter is that separation of lignin oil containing monomers and oligomers did not lead to improved performance of the coating. Therefore, the separation of lignin depolymerized fraction is not always necessary for the application in polymeric materials. Using the mixture obtained as such will have a tremendous benefit of reduced environmental impact and lower cost of production of said lignin oil compared with separated lignin oil fractions. I hope that this development will inspire more industrial and academic research on lignin oil mixtures used as such and that lignin separation would be employed when there is an actual benefit or necessity of its use.

The last major objective of this work is to study the relationship between the structure of the lignins obtained in depolymerization and fractionation processes and their properties, which affect their performance in real-life applications. Although this aim is addressed in all the chapters, it is the most pronounced in chapter 4. In that chapter, it was shown which structural features of lignin fractions lead to best antioxidant properties. In this chapter, the antioxidant properties of lignin were utilized to serve as an antioxidant additive in biolubricants, which was possible upon partial lignin esterification. The results of this research are highly relevant in the field of biolubricants given that the tribological properties of said biolubricants were improved or at least maintained, while thermo-oxidative stability was significantly improved. Low thermo-oxidative stability is the biggest downside of the use of vegetable oils in biolubricants, which can be overcome by incorporation of lignin-based additives.

The impact of this work on society is rather indirect. While society acknowledges the need to shift from the linear to the circular economy, it is not always straightforward how to implement this goal. That is why the work of scientists is important in addressing that need by showing the possible solutions on how to utilize biobased materials in everyday applications and what their performance is compared to the known materials. The boom in biobased economy is seen by the increasing amount of biorefinery projects utilizing biomass as a feedstock, which would lead to the increased availability of high-quality lignin fractions. Therefore, the circular economy aspect of lignocellulose biorefineries can be seen as the reduction of the amount of waste (for instance, residual biomass) by its reuse and upgrading, which can be achieved by integrating biowaste as a feedstock for fuels, chemicals, and materials.<sup>10-13</sup> The use of lignin-fractions originating from currently scaled-up processes contributes to their valorization in marketable solutions answers the urge to include biobased components in (polymeric) materials, which is contributing to

making biorefineries economy more efficient and circular. I hope that this work will help strengthen the knowledge bridge between academic research and industry, which would benefit society in the long term by making biobased products more available on the market.

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