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Editorial

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Editorial: From understanding the biological function of lignin in plants to production of colloidal lignin particles

Monika Österberg, Mika Henrikki Sipponen and Gunnar Henriksson

ABSTRACT: There is a paradigm shift going on in lignin utilization. High-volume materials and chemicals, as well as more engineered materials, like carbon fibers or active carriers, are currently being developed. However, in order to enhance lignin utilization in these and other value-added products, there is a need for better understanding of lignin function in woody tissues plus control over isolation, fractionation and other processing technology. Therefore, part of this issue is dedicated to articles about lignin. These range from lignin extraction to scale-up of the colloidal lignin particle production process. Focus is on fundamental understanding of interactions, a necessity for successful applications of this complex biopolymer.

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Due to recent trends in climate policies, there is an increasing demand for sustainable energy and raw materials. Thus, improved utilization of biomass feedstock is gaining attention. In biorefinery processes, carbohydrates are efficiently utilized as fibers for paper and board, as polymers in, for instance, cellulose derivatives, or for production of biofuels or biochemicals. In these processes, extracted lignin is generally burned during the chemical recovery system, producing energy and steam, but large volumes can be separated making the mills more efficient, since the recovery boiler is often seen as a bottleneck in the process. Thus, there is considerably more lignin available than needed for energy use only, and as a consequence, there is a renewed interest in lignin research.

Lignin valorization has been studied for decades and suggested applications have been e.g. carbon fibres, adhesives, emulsifiers and building blocks for chemicals (Kai et al 2017). Nevertheless, the existing commercial products are still scarce, besides the production of vanillin as a food grade flavour, and lignosulphonates for multiple purposes such as dispersing agents in e.g. cement. There are many reasons for the relative failure to find applications for lignin, one being the lack of competitive properties and cost of lignin products compared to existing solutions. This again could be due to the difficulty in extracting pure lignin with standard, well defined properties from black liquor or other biorefinery residues, but also due to the chemical complexity of lignin as such. In the articles in this issue these problems are tackled from various aspects.

Nature is usually superior to man in clever design. Hence, a natural way to unlock the potential of lignin is to

understand its role in plants. In the mini review by Henriksson (2017) the biological functions of lignin and how Nature has designed the polymer to achieve them, are discussed. Lignin's main functions in the plant are to provide water resistance to the cell wall and hinder its swelling in water, to make the cell wall stiff and glue the cells together. As additional functions Henriksson mentions lignin's role in hindering biological degradation (Henriksson, 2017). The possibility for lignin to covalently crosslink polysaccharides are suggested to be a key factor for how lignin functions.

The first obstacle to overcome for obtaining value-added lignin products is an efficient way of extracting the lignin from the biorefinery residues. The extraction method has a strong impact on the lignin properties and the desired properties varies depending on the intended application. This issue is addressed in the articles by Jääskeläinen et al (Jääskeläinen et al, 2017) and Durruty et al (Durruty et al, 2017). The kraft pulping process is today the largest industrial chemical process using wood as a material feedstock. The Lignoboost process is a method to extract part of the dissolved lignin from the black liquor before it enters the recovery boiler. The process is based on precipitation achieved by acidification of black liquor with carbon dioxide (Alen et al., 1979) After the precipitation the lignin is washed at acidic pH and recovered by filtration (Öhman, Theliander 2006; Öhman et al 2007). In the article by Durruty et al (2017) the effect of precipitation conditions, more specifically pH and xylan content, on the filtration efficiency is explored. They found that xylan had a detrimental effect on the filterability, while highly acidic conditions were preferable. Jääskeläinen et al (2017) on the other hand, describes a method to obtain carbohydrate-free and highly soluble lignin fractions by aqueous acetone evaporation fractionation. The solvent fractionation protocol separates lignin based on its solubility and it requires much less solvent compared to more commonly used methods, which is an economical and environmental advantage (Jääskeläinen et al, 2017).

Much of the lignin-related research focuses on the efficient removal of lignin from biomass in order to enhance properties of pulp or biofuel. In this field it is essential to understand the interactions between lignin and cellulose, hemicellulose or enzymes. It is well known that there is a fraction of recalcitrant lignin that is very difficult to remove during pulping (Ragnar et al, 2013). Herein, Giummarella et al (2017) discuss the interactions between cellulose and lignin and suggests that hemicellulose removal using hot-water extraction leads to close cellulose-lignin association, thus hampering the efficient lignin removal during pulping.

Lignin has been found to have unwanted impact on the action of cellulases during the enzymatic saccharification process. These effects include non-productive adsorption,

denaturation, inhibition and physical constraints for the enzyme to reach the cellulose (Pan 2008). Hence, a pretreatment step is usually employed prior to hydrolysis to overcome the detrimental effect of lignin. In the critical review by Sipponen et al (2017) these detrimental, and also reinforcing lignin-enzyme interactions, are discussed. They combine findings from fundamental interaction studies using various methods and approaches and make an attempt to summarize the main effects of different pretreatments on the lignin properties and consequently on the effect on enzymatic saccharification.

In addition to the above-mentioned effect of lignin on ethanol fermentation, the biodegradation of lignin is also central in the global circulation of carbon. Furthermore, a firm understanding of the mechanisms of lignin biodegradation might be an inspiration for the development of novel chemical methods for degrading and modifying lignin. Konstantopoulou et al (Konstantopoulou et al, 2017) have studied a panel of lignins prepared from different cultivars of wheat straw. They found that the structure appeared to be similar according to the methods they applied, but when they investigated the action of a number of culture filtrates from lignolytic bacteria, they found that there were large differences between the lignins in their susceptibility to degrade. This study underlines that there is still very much that remains to be studied, both in the fine structure of lignin, and in the action of lignolytic enzyme systems.

A relatively novel approach to lignin valorization is to prepare well defined spherical lignin (nano)particles (Lievonon et al 2016; Zhao et al 2016). This approach has two advantages. Firstly, the particle shape and size open up new application possibilities like drug or pesticide delivery (Yiamsawas et al 2014) or as functional substance in sun screen formulations (Qian et al 2015). Secondly, the well-defined shape and surface chemistry of the particles could solve the problem of heterogeneity and poor dispersibility/solubility of technical lignins. While the approaches to prepare colloidal or nanosized lignin described in literature to this date have been in laboratory scale (Zhao et al 2016), Leskinen et al (2017) demonstrate that the nanoprecipitation method in aqueous media can be scaled from a few milliliters to several liters. Scalable production processes are essential for these particles to find commercial use. Leskinen et al (2017) furthermore discuss the particle formation mechanism and show how various parameters affect the particle properties.

Further understanding of lignin self-association process is obtained from the article by Fritz et al (2017). They discuss the aggregation of dissolved lignin and lignin nanoparticles with respect to electrostatic or steric interactions. Overall, the articles published in the current issue address many timely issues regarding fundamental understanding of lignin properties, its role in and around cell walls, and also how it affects various biomass conversion processes. Scalable technologies for producing more homogeneous and well-defined lignin fractions in terms of molecular weight and functional groups, or in terms of producing spherical colloidal particles will ultimately lead to the development of new lignin-based applications and solutions.

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