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Lignin-first via reductive catalytic fractionation to make lignocellulosic biorefinery more sustainable

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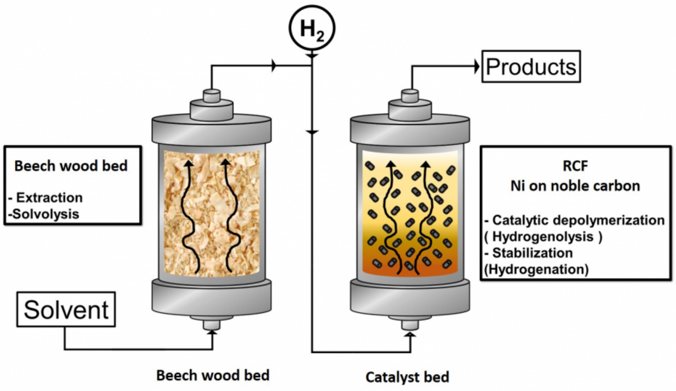
PURPOSE OF THE ABSTRACT

Intensive utilization of fossil resources for the production of energy, liquid transportation fuels, chemicals and plastics in the past century led to the current great environmental challenge of global warming.[1] Therefore, the transition towards carbon neutral societies with a circular bioeconomy independent of fossil resources is greatly needed to mitigate the influence of climate change. In this context and according to the United Nations Sustainability Plan and European Union 2050 vision (climate neutrality), lignocellulosic biorefineries is one of the main technologies to accomplish these plans. Traditional biorefinery usually focuses on making high value products from cellulosic biomass, whereas lignin fraction is considered as waste or utilized for energy recovery. To make biorefinery more sustainable and profitable lignin fraction should be valorized towards fine chemicals and/or materials. In this regard, lignin-first via reductive catalytic fractionation (RCF) approach was pioneered in batch systems yielding ~50% of monomers and low molecular weight aromatic oligomers.[2-4] Besides the excellent yield of monomers obtained, batch systems present some intrinsic disadvantages when compared with flow systems, such as the non-continuous cycle of operations, the difficult separation of products, catalysts and polysaccharides pulp, the mechanical disruption of pulp due to stirring, and the slow heating profile.[2-4] Additionally, batch systems required high reactor volumes for process intensification, which imply hazards and big spaces requirement.[4] Therefore we present here, the transformation of lignin-first via RCF from batch to continuous flow system using biomass bed (beech wood sawdust) and shaped Ni on noble carbon as a catalyst (catalyst bed) to bring this efficient approach a step forward towards real application (Figure 1). The use of flow system in lignin-first via RCF allows physical separation of the feedstock (wood bed) and catalyst bed, as the wood bed located upstream from the catalyst bed. Accordingly, in the first bed lignin is extracted from the lignocellulosic biomass and undergoes to solvolytic fragmentation, afterward in the downstream reactor lignin is catalytically depolymerized via hydrogenolysis and eventual non?stable double-bonds tails are stabilized over the hydrogenation catalyst. The de-coupled biomass and catalyst beds allow straightforward separation and different conditions applicable for the two RCF steps. In this approach, lignin extraction was found to be maximized in the first 4 h of time on stream (TOS), yielding a maximum cumulative monomers of 247 mg gKlason lignin-1 using MeOH as a solvent with respect to the use of 2-Methyltetrahydrofuran (MeTHF) that yielded 100 mg gKlason lignin-1 cumulative monomers, viz. Figure 2. In addition, extraction step temperature of 235°C and reductive fractionation temperature of 225°C was found to be the optimal condition for this process (Figure 2). Importantly, the catalyst was used for two cycles and a total time of 14 h of TOS without losing initial activity. In conclusion, the performing lignin-first strategy via RCF in continuous flow systems is a valid and promising possibility to make lignocellulosic biorefinery more sustainable and efficient.

FIGURES



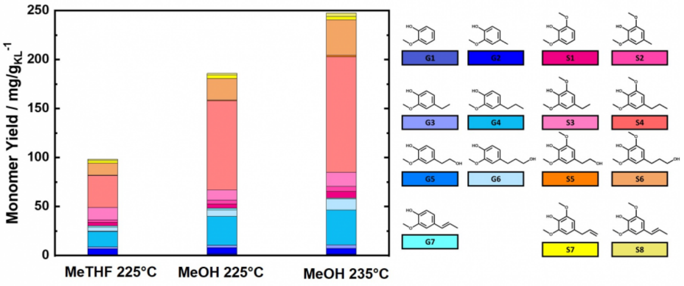


FIGURE 1

Figure 1

Representation of lignin-first via reductive catalytic fractionation of beech wood sawdust in a continuous flow system.

FIGURE 2

Figure 2

Monomer yield of RCF process after 10 h of TOS in function of the used solvent (MeTHF and MeOH); Reaction conditions: m35Ni/NDC = 10 g, mWood = 10 g, Textraction= 225°C, and 235°C Treduction = 225°C, pH2= 7.0 MPa, Qeduct = 1.0 mL min-1, QH2 = 48

KEYWORDS

Biorefinery | Lignin-first | Continous flow system | Heterogenous catalyst

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