ISGC 2022

N°584 / OC
TOPIC(s) : Biomass conversion / Homogenous, heterogenous and biocatalysis

# Controlled depolymerization - hydrogenation as innovative strategy for condensed tannin valorization.

## AUTHORS
Thijs VANGEEL / KULEUVEN, CELESTIJNENLAAN 200F - BUS 2461, LEUVEN
Bert SELS / KULEUVEN, CELESTIJNENLAAN 200F - BUS 2461, LEUVEN

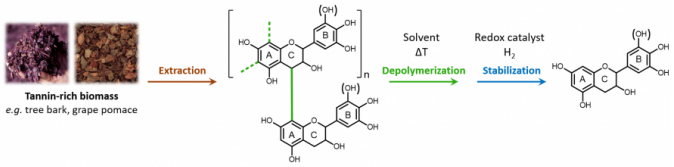
## PURPOSE OF THE ABSTRACT

Condensed tannins represent - after lignin - the largest source of renewable phenolics on earth [1]. They are present in important waste streams, such as tree bark from the forest industry and grape pomace from wineries, and thus constitute an interesting substrate for chemo-catalytic upgrading. In this contribution, we demonstrate a controlled depolymerization - hydrogenation strategy for converting these condensed tannins into their interesting monomeric constituents, i.e. monoflavanols.

Depolymerization of condensed tannins is typically carried out by acidic cleavage of the interunit bonds. This results in highly reactive carbocation intermediates that are prone to repolymerization reactions. Traditionally, these carbocation intermediates are stabilized through the addition of a strong nucleophile (e.g. thiol) - forming a nucleophile adduct - thus effectively avoiding repolymerization [2]. However, the very need for this sacrificial agent (the nucleophile) - often in large excess - has limited the use of this depolymerization strategy to analytical purposes only.

Inspired by the promising results obtained for lignin in the Reductive Catalytic Fractionation process [3] - during which lignin intermediates are stabilized via catalytic reduction - we set out to develop a similar strategy for condensed tannins. We demonstrate that condensed tannin interunit bonds can also be broken in neutral conditions at elevated temperature. Crucially, we show that the generated reactive intermediates can be stabilized via hydrogenation over a heterogeneous redox catalyst, thereby avoiding detrimental repolymerization. The outcome of this process is a high selectivity towards only one monoflavanol (and its stereoisomers), e.g. catechin and epicatechin.

## FIGURES




## FIGURE 1

Condensed tannin depolymerization - hydrogenation

Condensed tannins are depolymerized at elevated temperature, followed by the immediate reductive stabilization of the generated reactive intermediates.

## FIGURE 2


## KEYWORDS

Condensed tannins | Heterogeneous catalysis | Biomass | Polyphenols


## BIBLIOGRAPHY


[1] P.E. Laks, An overview of condensed tannin structure, in: R.W. Hemingway, J.J. Karchesy (Eds.) Chemistry and significance of condensed tannins, Springer, (1989) 131-136.

[2] S. Matthews, I. Mila, A. Scalbert, B. Pollet, C. Lapierre, C.L. Hervé du Penhoat, C. Rolando, D.M. Donnelly, Method for estimation of proanthocyanidins based on their acid depolymerization in the presence of nucleophiles, J. Agric. Food. Chem., 45 (1997) 1195-1201.

[3] T. Renders, G. Van den Bossche, T. Vangeel, K. Van Aelst, B. Sels, Reductive catalytic fractionation: state of the art of the lignin-first biorefinery, Curr. Opin. Biotechnol., 56 (2019) 193-201.