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The synergistic relationships between main chain-acting and debranching enzymes towards the efficient biodegradation of xylan

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PURPOSE OF THE ABSTRACT

Enzymatic utilization of lignocellulosic biomass for industrial applications has been proposed to require a wide spectrum of biocatalytic activities. Hemicellulose is a complex heteropolymer that confers rigidity on biomass structures and recalcitrance against biodegradation. Xylan in particular, mostly encountered in hardwood biomass, carries a variety of substitutions that promote molecular interactions with cellulose and lignin. Acetic acid, ferulic acid, arabinosyl and glucuronoyl residues are some of the most common substitutions that occupy positions O-2, O-3 or even O-4 of the xylopyranosyl residue (Xylp), and has been shown to inhibit enzymatic hydrolysis of the xylan main chain by xylanases and xylosidases. Therefore, the addition of accessory enzymes, such as acetyl-esterases, ferulic acid esterases, glucuronoyl esterases, α-L-arabinofuranosidases, α-glucuronidases and lytic polysaccharide monooxygenases, can help overcome the recalcitrance of hemicellulose by either removing the decorations of xylan or disassociating xylan from other biomass components and thus enabling xylanases to proceed with hydrolysis.

In this work, the synergistic effect between a number of accessory enzymes and a set of hemicellulose main-chain degrading enzymes has been examined, in order to gain some insight regarding the modifications of the biomass required in order to promote their activity. During hydrolysis of arabinoxylan-containing substrates, the removal of arabinofuranosyl residues was investigated using two arabinofuranosidases of distinct specificities, TtAbf43 and AnAbf51, in combination with xylanases of different glycoside hydrolase (GH) families. TtAbf43 removes the side-group from position O-3 of doubly arabinofuranosyl-substituted Xylps, whereas AnAbf51 hydrolyzes arabinose substitutions from positions O-2 or O-3 of singly substituted xyloses. The disassociation of glucuronoxylan form lignin moieties was investigated with the use of two glucuronoyl esterases of the carbohydrate esterase (CE) family 15, TlGE15 and AeGE15, by testing activity on the liquid and the solid fraction of the xylanase-hydrolyzed pretreated beechwood samples.

The activity of accessory enzymes on hemicellulose resulted to increased hydrolysis efficiency by the xylanases. During hydrolysis of polymeric arabinoxylan, TtAbf43 was able to unblock the xylobiohydrolase activity of TtXyn30A, while in the presence of both arabinofuranosidases the xylobiose release was further increased. During hydrolysis of pretreated wheat bran, TtAbf43 and AnAbf51 exhibited synergistic relationships with xylanases of the GH10, GH11 and GH30 families, where both xylanase and arabinofuranosidase activities were enhanced. Regarding the degradation of glucuronoxylan-lignin complexes, TlGE15 and AeGE15 exhibited activity both on the xylanase-derived hydrolysate and the residual biomass fraction. Overall data suggests that a cautious design of enzymatic cocktails, bearing in mind the composition of each substrate and the exact specific activities of the enzymes to be involved, could significantly promote biomass biodegradation. In this way, exploitation of residual biomass gains potential for the production of value-added carbohydrate-based chemicals, while minimizing the enzyme load of the treatment, contributing thus to the concept of circular bioeconomy.

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FIGURES

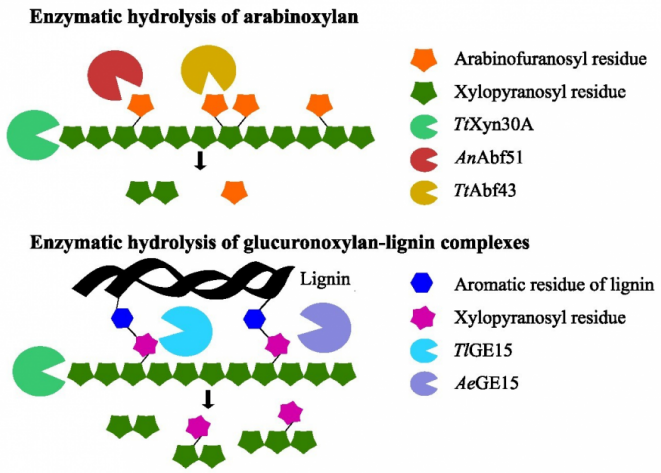

FIGURE 1

Enzymatic hydrolysis of arabino-and glucuronoxylan

Fig. 1. Enzymatic hydrolysis of decorated arabino- and glucuronoxylan

FIGURE 2

Acknowledgements

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KEYWORDS

biorefinery | lignocellulose decomposition | enzyme cocktails | xylan degradation

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