Developing a Targeted Mass-Spectrometry Platform for High-Throughput Characterization of Lignocellulosic Biomass Deconstruction

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Project Goal: This work aims to develop an analytical platform for high-throughput analysis of lignocellulosic biomass deconstruction. This platform combines surface-based mass spectrometry with bioconjugation chemistry to screen enzymatic and microbial activity toward lignin, cellulose, and hemicellulose degradation.

Lignocellulosic biomass is the most abundant raw material on the planet and is composed primarily of the cellulose, hemicellulose and lignin. Understanding how to deconstruct this complex material is critical to achieving a cost-competitive replacement for petrochemicals. Enzymes and microbes are capable of degrading this biomass in nature; however, the species, mechanisms, and pathways involved in deconstructing this material are vast and complex. To this end, we are developing mass spectrometry-based assays as a high-throughput platform to support the rapid development of minimal cocktails for lignocellulosic deconstruction. We present complementary developments in our mass spectrometry surfaces and the chemical probes used for targeted analysis of biomass degradation.

We present a new surface assisted laser desorption ionization method, Insulator Nanostructure Desorption Ionization Mass Spectrometry (INDI-MS). INDI-MS utilizes a self-assembling perfluoroalkyl silsesquioxane coating to achieve femtomolar sensitivity comparable to the hydrofluoric-etched Nanostructure Initiator Mass Spectrometry (NIMS) chips previously developed by our group. In addition to removing the etching step, this new method can also be integrated with photolithography to achieve sample self-desalting and enhanced performance.

To enhance our nanostructure-based MS detection of lignocellulosic biomass degradation products, our assay approach utilizes two chemical probe techniques to target activity toward the separate polysaccharide and aromatic components. A previously-developed perfluorinated tagging technique targets glycan products for rapid and reliable characterization of cellulose and hemicellulose degradation on our nanostructured surfaces. This approach has since been extended to characterize ligninase activity towards for β-O-4 type compounds. We show that unlike colorimetric assays used to study lignin decomposition, this lignin assay can provide unique and important information about specific bond cleavage reactions.

References

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