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One-Step Synthesis of N-Heterocyclic Compounds from Carbohydrates over Tungsten-Based Catalysts

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PURPOSE OF THE ABSTRACT

There is a growing desire to generate valuable chemicals and commodities from renewable resources such as biomass, for the benefits of the environment and the sustainable development of our society. Recently, research efforts have been made to obtain value-added N-containing chemicals from renewable biomass.[1-4] N-heterocyclic compounds such as pyrazines are highly valued chemicals with wide applications agricultural, pharmaceutical, cosmetic, etc. Currently, the manufacturing of these compounds is achieved via multiple transformation steps from fossil oils, which is not renewable and not very efficient. The objective of the abstract is to put forward a simple and efficient way to synthesis N-heterocyclic compounds from biomass resources via a one-step transformation strategy.[5]

Glucose is the most abundant renewable sugar that can be easily obtained from inedible cellulosic biomass. In this abstract, a one-step synthesis of N-heterocyclic products such as 2-methyl pyrazine from glucose and other carbohydrates is described, via an effective catalytic system using aqueous ammonia as the nitrogen source. This new method offers a competing synthetic route to the conventional method. Under optimal conditions, 2-methyl pyrazine (MP) and 4(5)-methyl imidazole (MI) were produced within 15 min at 180 °C with combined yields of around 40%. While the formation of 4(5)-MI was identified as a noncatalytic process, the yield of 2-MP was highly influenced by the presence of catalysts. Nearly 3-fold yield enhancement of 2-MP was achieved over several tungsten-based catalysts. Control experiments, isotope-labeling tests, ESI-MS, and NMR analysis revealed that the formation of 2-MP follows a fragmentation mechanism, with beta-D-glucopyranosylamine as an important intermediate. Besides, small W2-W4 tungsten clusters were plausibly assigned as the catalytically active species facilitating both glucose fragmentation and the subsequent cyclization reaction to generate pyrazine rings. The catalytic system is applicable to other monosaccharides and some disaccharides. The work points out new possibilities on the sustainable production of N-heterocyclic compounds from biomass resources through simple, catalytic routes.

FIGURES

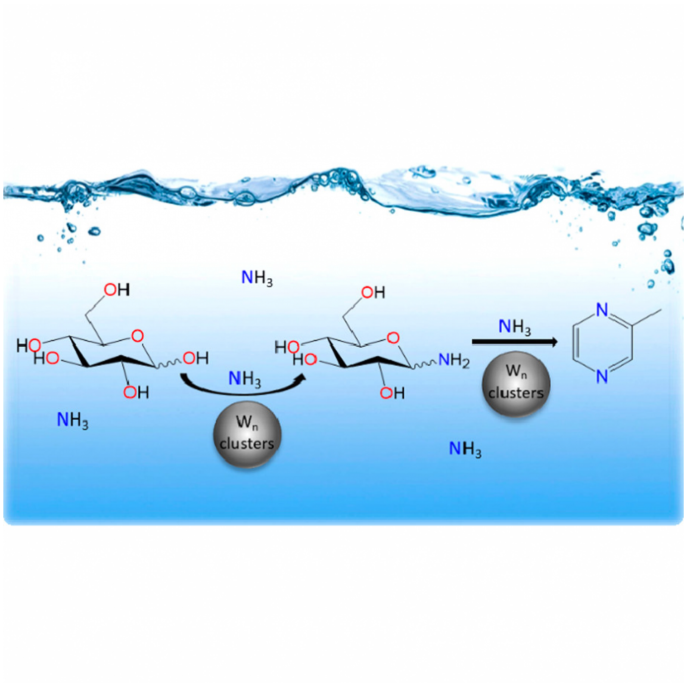

FIGURE 1

Fig.1

Tungsten-catalyzed, one-step conversion of glucose into N-heterocyclic compounds

FIGURE 2

KEYWORDS

Biomass utilization | Homogeneous catalysis | Renewable chemicals | N-heterocyclic compounds

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