492d Probing Higher Polyol Hydrogenolysis through Stereoisomer Reactions

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Not only does higher polyol hydrogenolysis to ethylene glycol and propylene glycol have potential commercial implications, the reaction provides an interesting system to develop more fundamental understanding on the use of supported metal catalysts for the conversion of carbohydrate-derived molecules. However, the production of the desired glycols requires competing catalytic activity for C-C and C-O scissions and dehydrogenation/hydrogenation leading to significant reaction complexity. This complexity has limited most of the previously reported results to being largely empirical. Development of more fundamental understanding for this reaction will necessitate utilizing a number of different characterization and reaction study approaches. We have investigated the use of stereoisomer reaction studies to develop insight into the important mechanisms involved in the reaction. Nine polyols ranging from three to six carbons were reacted under hydrogenolysis conditions. The resulting products were used to determine the relative importance of the retro-aldol and decarboxylation mechanisms in C-C scission reactions. Information about the relative dehydrogenation rates along the polyol as well as a selectivity map describing the carbon flux in the overall reaction was found. Finally, the implications of sulfur addition on this selectivity map were determined by comparing reaction results for the polyol stereoisomers.