IMIDAZOLIUM CATALYSTS FORMED BY AN ITERATIVE SYNTHETIC PROCESS AS A MODEL SYSTEM FOR CHEMICAL EVOLUTION

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Abstract: Processes exhibiting diversity and selection would have been necessary to promote chemical evolution on early Earth. In this work a model process was developed using non-kinetic selection to synthesize and isolate small molecule imidazolium catalysts. These catalysts were purified by affinity chromatography, and recycled back into the process forming a product feedback loop. In dimethylformamide the catalysts activated the coupling of formaldehyde to short chain sugars. This sugar mixture was reacted with aniline, acetic acid, and paraformaldehyde to generate new catalysts. Thus structural diversity was produced through non-selective, multi-component synthesis. Applying sequential dilution-reaction-purification cycles we demonstrated that this process can function independently of starting catalyst. Over 4 process cycles, the initiator catalyst is effectively diluted out as a new catalyst population emerges to take its place. This system offers an alternative viewpoint for chemical evolution via the generation of small molecule organocatalysts.

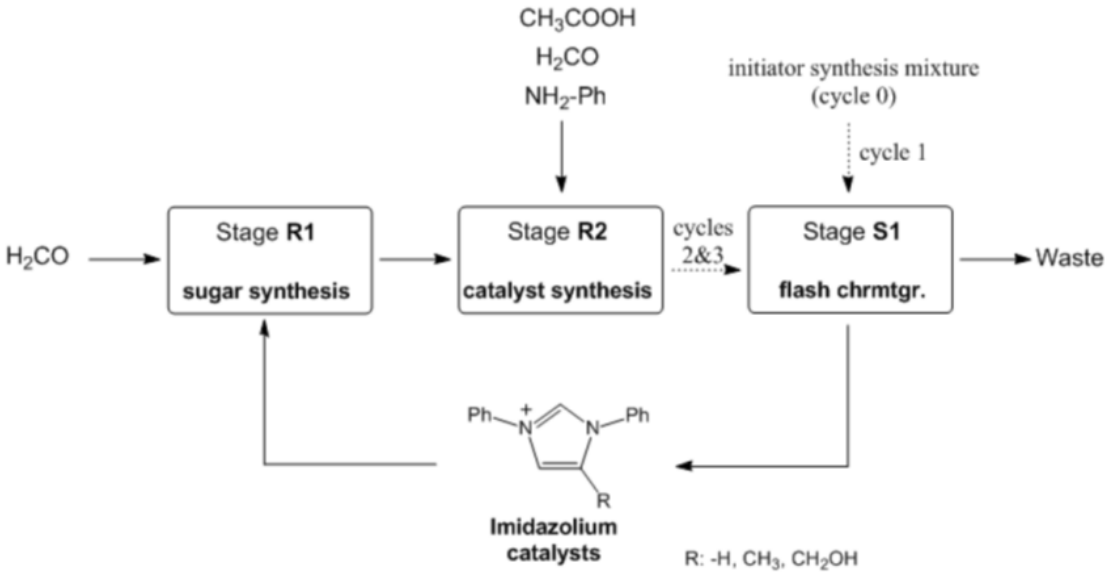

Figure 1. Process Flow Diagram illustrating the three-stage synthesis and purification of imidazolium catalysts from formaldehyde (H_2CO), aniline (NH_2-Ph), and acetic acid (CH_3COOH). Stage R1 is the catalytic conversion of formaldehyde to various sugars in dimethylformamide. Stage R2 is the multi-component reaction of these sugars with aniline, formaldehyde, and acetic acid. Stage S1 is the purification of imidazolium catalysts by flash chromatography. The process is initiated using dihydroxyacetone as a standard sugar to form primer catalysts in cycle 0. The primer catalysts were diluted, then recycled back into stage R1 to generate new catalysts in subsequent process iterations.Thus creating a product feedback loop.