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Induction of asymmetry in formose reaction

I. Myrgorodska¹ and S. P. Fletcher¹

¹Department of Chemistry, University of Oxford. Chemistry Research Laboratory, 12 Mansfield Road, Oxford, OX1 3TA, UK

* iuliia.myrgorodska@chem.ox.ac.uk

The autocatalytic transformation of formaldehyde and glycolaldehyde to carbohydrate building blocks, also known as formose reaction, is a cornerstone of prebiotic chemistry. In fact, it has been argued that formaldehyde may be the only one carbon C-, H- and O-containing molecule capable of generating complex organic compounds for the origin of life.[1] Sugars are key components of DNA, RNA and the surfaces of cells, where they perform a wide variety of complex regulatory and communication roles. Recently, sugar molecules including ribose have been detected in interstellar ice analogues suggesting the occurrence of photochemically initiated formose-type reactions,[2] thus demonstrating that carbohydrate can be formed under a wider range of conditions than believed. Even though formose reaction in general is robust, it has numerous limitations including low yields, absence of chemo- and stereoselectivity. The problem of enantioselectivity has been previously addressed by Breslow[3] and Pizzarello[4] as they used amino acids and peptides to generate asymmetry in C3 to C5 sugars.

In the present work we investigated influence of different chiral species, including amino acids and sugar derivatives, on the enantioselective outcome of the formose reaction. These data have been used to gain better understanding of the reaction network. Furthermore, we examined possibility of generating formose reaction within prebiotically relevant systems such as chiral vesicles. Understanding the relationship between key intermediates of the formose reaction may inform the design of an asymmetric autocatalytic reaction.

References: [1] Weber A (2002) *Origin of Life and Evolution of Biosphere* 32:333–357. [2] Meirnet C. et al. (2016) *Science* 352:208–212. [3] Breslow R. and Cheng Z. L. (2010) *PNAS*, 107:5723–5725. [4] Weber A. L. and Pizzarello S. (2006) *PNAS*, 107:5723–5725.

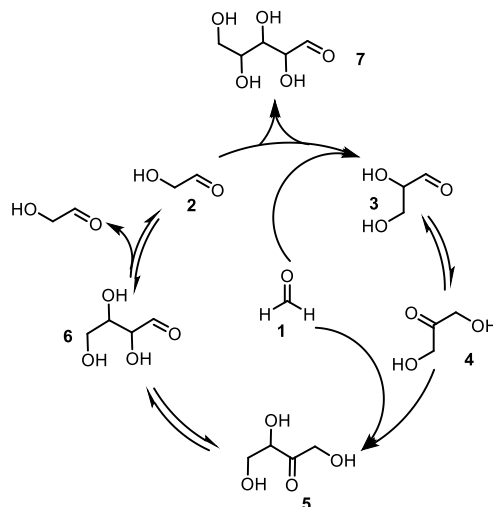


Figure 1 The first stage of formose reaction involves condensation of formaldehyde **1** with glycolaldehyde **2** to yield chiral aldehyde **3**. Aldol condensation of **3** and **2** produces pentoses, while condensation of two molecules of glycolaldehyde **2** results in tetroses **6**. Glyceraldehyde **3** isomerises to dihydroxyacetone **4**. Condensation of **4** with **1** produces erythrulose which in turn can isomerise to **6**.