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Title:	Insights into the kinetics and mechanism of spermine (base)-catalyzed D-fructose interconversion to low-calorie D-allulose
Authors:	Mahala, Sangeeta (/jspui/browse?type=author&value=Mahala%2C+Sangeeta) Devi, Bhawana (/jspui/browse?type=author&value=Devi%2C+Bhawana)
Keywords:	D-fructose spermine (base)-catalyzed D-allulose
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Abstract:	The low-calorie D-allulose as a food ingredient has the potential to reduce lifestyle diseases (obesity and diabetes). The biological method widely followed for its production represents an expensive sweetener. Unfortunately, the chemical method is seldom used, probably due to the lack of understanding of the catalytic mechanism. Here, we report the chemical kinetics and mechanism of fructose into allulose by using spermine, a weak-base catalyst. The kinetic results of the homogeneous catalysis elucidate that fructose has a higher affinity to develop an equilibrium relationship with the formed side products (glucose and mannose) via competitive C2 isomerization rather than with allulose via C3 epimerization. The ready transformation of fructose to side products under a basic condition shifts the equilibrium towards fructose, resulting in a reduced allulose formation (8% yield and 21% selectivity). The proposed reaction mechanism unveils the allulose synthesis obeying the LdB-AvE principles using the 2,3-enediolate (however, its development is dependent on the degree of both forward fructose and reverse allulose transformations), while the side products using a single 1,2-enediolate. Of the possible synthesis pathways that spermine could promote, the proton transfer pathway requiring a 27% lower energy than 1,2-hydride transfer is feasible, ascribed to the in-situ synthesis of fewer intermediary molecules. Our findings offer new insights into acid/base-mediated fructose transformation in water to design an efficient catalytic system to produce allulose in a shorter time.
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