

Efficient hydrolysis of glucose-1-phosphate catalyzed by metallomicelles with histidine residue

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Phosphate esters play an important role in genetic information transfer, cell signal transduction, energy transmission and metabolic processes of living beings. Efficient catalytic hydrolysis of phosphate esters is still an attractive and challenging problem. Here, a new 2-amino-N-dodecyl-3-(1H-imidazol-5-yl)propanamide (L²) surfactant was synthesized and its metallomicelles of La³⁺, Cu²⁺, Co²⁺, and Zn²⁺ complexes were used as mimic metalloenzymes to catalyze the hydrolysis of glucose-1-phosphate (G1P) in a buffer solution at 35 °C. The metallomicelle systems can efficiently catalyze the hydrolysis of G1P. The rare-earth metallomicelle LaL² has the highest catalytic activity compared with those of the transition metal micelles CuL², CoL², and ZnL². Different association behaviors of metallomicelles and substrate G1P were proposed. The imidazole group might accelerate the hydrolysis by activating H₂O associated with the metal into a metal–OH⁻ group. A possible catalytic mechanism was also discussed.

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Introduction

 α -D-Glucose-1-phosphate (G1P) is a product of intracellular oligosaccharide or polysaccharide degradation (Campbell et al., 1952). It is also a precursor for the synthesis of polysaccharides and nucleotides (Rashid et al., 2004). The backbone of RNA and DNA is constituted of sugar molecules and phosphates linked by the 3',5'-phosphodiester bond. Intensive investigation of the catalytic formation and shear of the phosphate-sugar ester bond is essential for the understanding of energy transfer, expression of genetic information and sugar metabolism, as well as for diabetes and tumor therapy. Therefore, G1P was selected as a model of RNA and DNA in the study of the cleavage of the bond between sugar and phosphate. However, various researches have focused on the catalytic hydrolysis of phosphate-sugar because of the stability of the phosphate-sugar ester bond. Recently, Olsson et al. (2010) investigated the hydrolysis of G1P promoted by goethite. Unfortunately, the catalytic activity of goethite is not high, and the biocompatibility of the heterogeneous catalytic system is also poor. Metallomicelle, as a new self-assembling supramolecular system, can mimic the active center and hydrophobic microenvironment of the metalloenzyme and has thus attracted considerable interest in recent decades (Gellman et al., 1986; Jiang et al., 2005; Bhattacharya & Kumari, 2009; Desbouis et al., 2012; Xie et al., 2013). As an excellent multifunctional system, metallomicelles have also been widely applied in reactions catalysis (Ye et al., 2008; Boudou et al., 2006) and new materials preparation (Seidel et al., 2007; Li et al., 2012; Sasidharan et al., 2012). Characteristics of a metallomicelle are dominated by the properties of the metal ion and functional groups. Histidine residue plays the key role in the enzyme-catalyzed reactions. Imidazole of the histidine residue can form a hydrogen bond with the substrate and the H proton of the imidazole can be rapidly transferred between the two

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