The crustal uplift determined at the Jakobshavn glacier (West Greenland) using ATM and GPS data

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Publication date:
2012

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

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Didehydroxylation – a new approach for conversion of biomass

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INTRODUCTION

The available amounts of fossil fuels are rapidly diminishing while the consumption is continuously growing. Thus it is necessary to find a new source of fuel. Recently the focus on using biomass as a renewable source has increased, but while the industrial production of second generation biofuels is the best possible scenario, it does not come without challenges. First and foremost, developing a cost-effective process for conversion of biomass is a necessity. One challenge is to decrease the ratio between oxygen and carbon in the biomass in order to increase the energy density. In this project a series of initial didehydroxylation experiments has been carried out to determine the reactivity of different styrene diols as a function of their substitution pattern.

DIDEHYDROXYLATION

The osmium-catalyzed dihydroxylation of an alkene using OsO₄ is a well-studied reaction whereas the didehydroxylation of a vicinal diol using Re₂(CO)₁₀ just recently attracted attention.¹

\[
\text{HO-CH}_2\text{OH} + \text{Re(OH)O}_2 \xrightarrow{\text{heat}} \text{H}_2\text{O} + \text{Re(OH)O}_3
\]

The reaction could occur via mechanism similar to the one for the dihydroxylation, just reverse. First the rhenium catalyst is oxidized to its active form, Re⁶⁺(OH)O₂ (fig.1), after which it reacts with the two hydroxy groups of the diol, forming an intermediate rhenium complex while releasing two water molecules. The alkene is then released, leaving rhenium in oxidation state VII. Rhenium is then reduced back to Rh⁴⁺ by oxidation of 3-octanol.

HAMMETT STUDY

The reactivity of the different para-substituted diols (fig.2) has been investigated by determining the relative reactivities of the didehydroxylation reactions. The reactions have been followed using both GC and HPLC as well as ^1H-NMR in order to follow both the formation of the product and the disappearance of the reactant. Several different reaction conditions have been examined, but all used 3-octanol as a reductant for rhenium, making the reaction catalytic. The influence of the different para-substituents on the reactivity and reaction mechanism of the didehydroxylation can then be evaluated by the construction of a Hammett plot.

\[
\begin{align*}
\text{HO-CH}_2\text{-} & \quad \text{R} \quad \text{HO} \\
\text{OH} & \quad \text{OH} \\
\text{Re}_2(\text{CO})_{10} & \quad (2 \text{ mol/\%})
\end{align*}
\]


The results obtained for the predicted crustal uplift for KAAS is 11.62 mm/yr while the observed value was 16.321 mm/yr, for ILAS 1.74 mm/yr and 1.53 mm/yr, for QEAS -0.189 mm/yr and 1.15 mm/yr. That being a difference of 4.701 mm/yr is found for KAAS, 0.21 mm/yr for ILAS and 1.339 mm/yr for QEAS. The uncertainties associated both with the ATM and GPS results are 0.8 mm/yr for ATM and 0.5 mm/yr for GPS. The total ice mass loss in km² of water predicted from the ATM data concerning the Jakobshavn area is -88.815 between 2010 and 2005 and -83.599 between 2005 and 1997.

It seems fair to state that the differences, between the predicted and observed rates, may also be due to the fact that not all the errors have been taken into account when computing the observed results and also due to the fact that, perhaps, ice is melting in Greenland much faster than predicted.

REFERENCES