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Review Article

A review of catalytic aqueous-phase reforming of oxygenated hydrocarbons derived from biorefinery water fractions

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ABSTRACT

Aqueous-phase reforming (APR) of oxygenated hydrocarbons is a process for the production of hydrogen and light alkanes. The reactants of APR remain in liquid phase during the reaction avoiding an energetically demanding vaporization-step compared to processes such as steam reforming (SR). Furthermore, low reaction temperatures thermodynamically favour the formation of hydrogen with low carbon monoxide content. Therefore, APR has been recently considered as a promising route to upgrade organic compounds found in biorefinery water fractions.

Aqueous oxygenated hydrocarbons are reformed at low temperatures (200–250 °C) and high pressures (15–50 bar), typically with platinum- and nickel-based catalyst. In addition, metal combinations of these and other metals have been applied to enhance the catalyst performance. Alumina has been extensively used as catalyst support in APR. Nonetheless, other oxides and carbonaceous materials have been applied to improve the stability of catalysts.

Hydrothermal conditions and high pressure present operational challenges that hinder the development of aqueous-phase reforming. However, low yields of desired products and fast catalyst deactivation constitute the main barriers. To maximize the APR effectiveness, the optimization of operation conditions and more active and durable catalysts are required.

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Introduction

The transition from the fossil-based to a bio-based economy demands the replacement of conventional, unsustainable processes. Biorefineries represent an alternative to produce chemicals, fuels, power and heat from renewable lignocellulosic biomass. Representative examples of biorefineries include thermochemical processes such as gasification and pyrolysis [1]. The target products of these processes are frequently accompanied by an aqueous fraction with organic compounds, for instance oxygenated hydrocarbons. The disposal of these water fractions entails costly conditioning treatments. Additionally, misusing the organic compounds contained in the water fractions decreases the feedstock-usage efficiency compared to the case in which these organic compounds were upgraded to valuable products. The value of all the product streams should be maximized to increase the production efficiency of biorefineries.

Hydrothermal processes have been applied to convert lignocellulosic biomass or intermediate products from biomass processing with high water content into biochemicals and biofuels. These processes are conducted in liquid phase applying temperatures in the range of 100–600 °C and pressures between 1 and 400 bar [2,3]. An advantage derived from these conditions is the suppression of an energetically demanding vaporization-step. Hydrothermal processes can be classified into two categories, sub- and supercritical technologies, based on the critical point of water [2]. Water reaches the critical point at 374.2 °C and 221.2 bar. Among the processes that apply subcritical conditions [4], this work focuses on aqueous-phase reforming (APR). In this type of reforming, hydrogen and carbon dioxide are produced among other gaseous and liquid products from organic components in aqueous phase. Hydrogen is a renewable energy carrier that can be converted to electricity and heat. In

addition, hydrogen is utilized in the hydrotreatment of sustainable biomass resources for biofuel production. The environmental advantages of producing non-fossil hydrogen are well known. Accordingly, hydrogen is commonly considered as the main product of APR.

Oxygenated hydrocarbons such as sugars and alcohols were the feedstock first proposed in 2002 by the group of Dumesic to produce hydrogen applying platinum catalysed aqueous-phase reforming. Cortright et al. [5] studied the performance of platinum-based catalysts to convert aqueous C:O = 1:1 oxygenated hydrocarbons into hydrogen. Few years after this first publication, Virent Energy Systems, Inc., with Cortright as Executive Vice President, claimed to be commercializing the Aqueous Phase Reforming process for the production of hydrogen and gaseous alkanes from glycerol and sorbitol [6]. Today, Virent, Inc. [7] pursues to scale up its patented technologies which apply aqueous-phase reforming to plant-based sugars to produce hydrocarbon products, including gasoline, diesel, jet fuel, and chemicals.

During the past years, the amount of studies focused on APR to produce hydrogen and alkanes has increased. Researchers have examined feedstock components and concentrations, catalysts with different active metals, promoters and supports, variations in reaction conditions, and a few reactor designs. However, the aqueous-phase reforming process still remains in laboratory scale. Further research is needed to optimize the process productivity and launch APR to scales closer to commercialization.

This work reviews the state of catalytic aqueous-phase reforming of oxygenated hydrocarbons. First, aqueous-phase reforming is introduced and compared with other reforming technologies. Second, various parameters to evaluate the APR performance are presented, and discussed. Third, the performance of APR is evaluated considering different feedstock, reaction conditions, and catalysts. Additionally, this paper analyses the reactor technologies

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