

Pt/Ga₂O₃ catalysts of selective hydrogenation of crotonaldehyde

E. Gebauer-Henke^a, J. Grams^a, E. Szubiakiewicz^a, J. Farbotko^a, R. Touroude^b, J. Rynkowski^{a,*}

^a Institute of General and Ecological Chemistry, Technical University of Łódź, ul. Żeromskiego 116, 90-924 Łódź, Poland

^b LMSPC, UMR 7515 du CNRS, ECPM-ULP, 25 rue Becquerel, 67087 Strasbourg Cedex 2, France

Received 27 February 2007; revised 11 June 2007; accepted 17 June 2007

Available online 27 July 2007

Abstract

Hydrogenation of crotonaldehyde in a gas phase at atmospheric pressure over Pt/Ga₂O₃ catalysts was studied. Two types of platinum precursors [Pt(acac)₂ and H₂PtCl₆] and two gallia supports (α -Ga₂O₃ [60.7 m²/g] and β -Ga₂O₃ [2.2 m²/g]) were used for catalyst preparation. The catalyst 5 wt% Pt/ α -Ga₂O₃ prepared from Pt(acac)₂ precursor showed very high and stable selectivity to crotyl alcohol (91% and 63% at 10% and 70% crotonaldehyde conversion, respectively). Because Pt-supported catalysts, showing a high selectivity, usually have relatively low activity, the most interesting finding of this study is that the use of gallium oxide as a support for platinum makes it possible to obtain catalysts with significantly increased C=O bond hydrogenation selectivity while maintaining high activity.

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Keywords: Platinum; Selective hydrogenation; Crotonaldehyde; Gallium oxide; Unsaturated alcohol

1. Introduction

Selective hydrogenation of α,β -unsaturated aldehydes to unsaturated alcohols has been a subject of many investigations because of its great interest as an important step in the preparation of various fine chemicals and applications of unsaturated alcohols in the flavor and fragrance industries and in pharmaceutical manufacturing [1]. The challenge lies in the selective hydrogenation of the carbonyl bond while keeping the olefinic bond unaffected. This can be achieved using stoichiometric amounts of reducing agents, such as hydrides (e.g., AlLiH₄, NaBH₄); however, such methods are useful only for a small-scale production because they involve costly chemicals and are hazardous to the environment.

The development of specific catalytic hydrogenation processes can replace the reduction using metal hydrides, which is in agreement with one of the 12 principles of green chemistry: “Catalytic reagents (as selective as possible) are superior to stoichiometric reagents.” Catalysis offers advantages over stoichiometric reactions in terms of both increased selectivity and energy minimization. The specificity of catalysts favors the

reaction to a preferred product, whereas the amount of undesired products is decreased. The amount of energy required for a given transformation is also reduced as the catalysts decrease the activation energy of the reaction [2]. Consequently, hydrogenation processes based on heterogeneous catalysis have been developed [3]. Hydrogenation of α,β -unsaturated aldehydes to unsaturated alcohols is difficult to realize, especially under heterogeneous catalysis conditions, due to the much greater susceptibility to hydrogenation of the C=C double bond (578.8 kJ/mol) compared with the C=O double bond (705.8 kJ/mol) for both thermodynamic and kinetic reasons [4].

The reaction of hydrogenation of α,β -unsaturated aldehydes may occur in different ways, as shown in Fig. 1. α,β -Unsaturated aldehydes may be hydrogenated to the most desired product, unsaturated alcohols (step 1), as well as to other saturated products, butanal (steps 2 and 5) and butanol (steps 3 and 4).

The selectivity to unsaturated alcohols depends on various factors, such as the structure of aldehydes, the nature of catalysts, and the presence of additives, as well as the reaction conditions [5]. Previous studies have shown that unpromoted metals have specific selectivities to unsaturated alcohols. For example, iridium and osmium are rather selective; palladium, rhodium, and nickel are unselective or slightly selective; and

* Corresponding author. Fax: +48 42 631 31 28.

E-mail address: jacryn@p.lodz.pl (J. Rynkowski).

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