Guerbet Reaction

In subject area: Chemistry

The Guerbet reaction is a tandem multi-step process involving dehydrogenation, aldol condensation, crotonization, and hydrogenation reactions, used for synthesizing branched monoalcohols. It is a valuable method for producing industrial compounds like plasticizers, detergents, and bioalcohols, with the potential for further conversion into branched hydrocarbons for aviation fuels.

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Reactions and Mechanism

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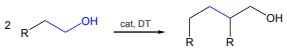
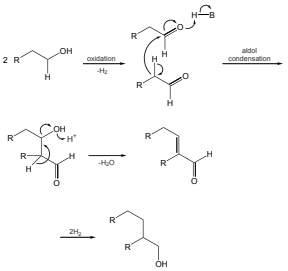
Catalytic conversion of a primary aliphatic alcohol into its β-alkylated dimer alcohol with loss of one equivalent of water.



Reaction



Mechanism

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2.3 Guerbet reaction

The tandem multi-step Guerbet reaction (involving dehydrogenation/aldol condensation/crotonization/hydrogenation reactions) is an attracting process for the synthesis of branched monoalcohols, value added compounds for industrial production of plasticizers and detergents, including the synthesis of bioalcohols (Fig. 10)[80,81]; further hydrodeoxygenation of the obtained branched alcohols enables the production of branched hydrocarbons, components of aviation fuels.[82]

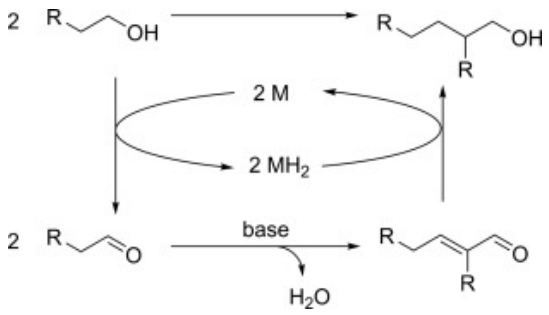

Fig. 10. Schematic representation of Guerbet reaction.

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Although catalysts consisting of a basic metal oxide (such as MgO)

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3.5 The Guerbet reaction

With the wide availability of bio-ethanol from first generation biorefineries, the direct conversion of ethanol into higher alcohols, *commonly known as the Guerbet reaction*, is a much-investigated transformation.[115–118] The simplest of the Guerbet alcohols is 1-butanol (Fig. 8), a molecule which is notably non-hygroscopic, non-corrosive, and of overall low toxicity.[119] From a fuel perspective, 1-butanol makes sense as it has an energy density of 29.3 MJ L^{-1}, which comes much closer to the one of gasoline (34.2 MJ L^{-1}) than does ethanol (24 MJ L^{-1}).[115] In this respect, it's also noteworthy that 1-butanol is fully compatible with gasoline engines as well as the existing fuel pipeline infrastructure. Butanol is fully miscible with gasoline in all ratios, and in the USA, it can be legally added to gasoline in up to 11.5%. It is also noteworthy that its lower vapor pressure and higher flash point makes it intrinsically safer to handle than gasoline.[119] Butanol also holds vast potential as a commodity and platform molecule. In this respect butyraldehyde and butyric acid are most noteworthy, while they have both mainstream applications in the solvent, polymer and specialty chemical markets. Also, butenes and butadiene hold additional opportunities as fuels and in the synthetic elastomers' markets.[119] Currently, butanol is mainly produced by the oxo process, which encompasses two steps notably (1) the cobalt/rhodium catalyzed hydroformylation of

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Gustavo Metzker, ... Mauricio Boscolo

3.3 Other relevant products indirectly obtained from ethanol

Although ethanol can be used as starting material for several chemical reactions aiming to produce high value-added chemicals, these products can also be employed as reagents to obtain organic compounds that are not accessible directly from ethanol. Among the GR derived products from ethanol, 1-butanol and its isomers deserve attention as carbon sources to produce valuable chemicals.[157–159] For example, synthesis of aromatic compounds from isobutanol by successive dehydration and aromatization reaction using zeolites (Si/Al = 13.4–34.3) as catalysts were studied.[160] The authors reported BTX compounds (benzene, toluene, and xylene) with yields ranging between 33% and 35% using a pure acid zeolite. Additionally, it was shown that the insertion of Zn on the zeolitic (2.3–5.1% wt) increased BTX yields by up to 52%.

Hydrocarbons obtained from 1-butanol using SiO_2-Al_2O_3 as catalysts were also reported.[161] The reaction was conducted under the vapor phase using a continuous down-flow fixed-bed reactor at atmospheric pressure and N_2 as a carrier gas. The results showed an excellent 1-butanol conversion (up to 96%) and C_3 and C_4 hydrocarbons selectivity (between 63% and 81%) at 250 °C. Under this condition, C_5–C_{12} hydrocarbons can be obtained with

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Synthesis of ethanol and its catalytic conversion

2019, Advances in Catalysis

Jifeng Pang, ... Tao Zhang

3.2.2.3 HAP catalysts

HAP materials, typically calcium-HAP, are well-known minerals used in the biological field. Recently, the interest in this material was extended to the field of catalysis, particularly as bifunctional catalysts or supports. The stoichiometry of calcium-HAP shows a molar Ca/P ratio of 1.67, i.e., $Ca_{10}(PO_4)_6(OH)_2$. Interestingly, the number of acid and base sites in HAP can be tuned through the substitution of calcium or phosphate ions, which shows high potential for alcohols conversion *(433,434)*.

Besides used in different condensation reactions, HAP has been tried in ethanol upgrading *(435,436)*. Tsuchida et al. tried various oxide catalysts, and found nonstoichiometric HAP with a Ca/P molar ratio of 1.64 selectively catalyzed ethanol to butanol at atmospheric pressure. At 573 K and a contact time of 1.78 s, ethanol conversion and butanol selectivity were 14.7% and 76%, respectively, with high carbon alcohols as co-products *(437)*. By changing reaction conditions (> 673 K) and feedstocks (different alcohols including ethanol, 1-butanol, 1-hexanol, 2-ethyl-1-butanol, 1-octanol and 2-ethyl-1-hexanol), gasoline with a research octane number of 99 was also obtained. The Guerbet reaction and the Lebedev reaction were demonstrated to be responsible for the C—C formation, followed by dehydrogenation and dehydration to aldehydes and olefins *(438)*. The same group

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Review article

Base metal-catalyzed alcohol C–C couplings under hydrogen transfer conditions

2018, Tetrahedron Letters

Yuan Cai, ... Shi-Liang Shi

C-Alkylation of alcohols

In place of carbonyl compounds, alcohols can also take place C-alkylation at the β-position through hydrogen transfer processes, which is an efficient method to synthesize more diversified alcohols. The earliest examples of the homocoupling of primary alcohols, namely the Guerbet reaction, were reported in 1909 (Scheme 15).[20] Guerbet reactions catalyzed by transition metals are normally believed to undergo through HT pathway involving twice dehydrogenation and twice hydrogenation. In 2012, Sun and coworker reported a novel iron-catalyzed β-alkylation of alcohols (Scheme 16).[21] Using commercially available ferrocenecarboxaldehyde as the catalyst and in the presence of a catalytic amount of base, the upgraded secondary alcohols can be obtained with 54–97% yield. When coupling secondary aliphatic alcohols with a primary aliphatic alcohol, higher temperatures (>180 °C) and neat conditions were needed. For the mechanism, the carbonyl moiety in the iron catalyst is believed to assist hydride transfer from alcohol substrates to metal center through a hemiacetal intermediate. The primary and secondary alcohols thus are oxidized to the corresponding aldehyde and ketone, followed by a base-mediated cross-aldol condensation and double reduction of the resulting enone by iron hydride to produce the desired upgraded alcohol products.

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Review article

Energy • Mechanistic biology

2015, Current Opinion in Chemical Biology

Aron Deneyer, ... Bert F Sels

Chemo- and biocatalytic tool box: the chemist's view

In order to accommodate a future transition to renewable carbon sources, development of new process technologies is crucial. Potential biomass feedstocks are sugar-based materials (for example hemicellulose and cellulose), lignin and glycerides, illustrated on the left-hand side of Figure 2. Their highly functionalized chemical structure and defined carbon skeleton are immediately apparent [17••]. Hence, techniques to remove the functional groups and adapt the C-number are necessary. In this way, a myriad of alkane structures with various properties can be realized. A few relevant examples are presented on the right-hand side of Figure 2. This paper, and more specific this paragraph, mainly focusses on a mild and selective chemocatalytic approach to obtain alkanes from biomass [18–20]. Figure 2 thus depicts an overview of the chemocatalytic techniques that can be used for the production of alkanes starting from glycerides, lignin and sugar-based materials. Furthermore, valuable cooperations between biological and chemical techniques are included, for example the valorization of biological conversion products like carboxylates by means of ketonization [21–24] or alcohols by coupling through a Guerbet reaction [25,26••,27,28]. Other conversion technologies, like (hydro)pyrolysis [29•,30] and

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Butene, Furfural, Condensation, Hydrogen, Layered Double Hydroxides, Dehydrogenation, Mixed Metal Oxide, Aldol Condensation, Porosity.

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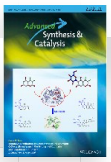

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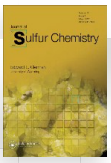

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