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Preparation of gold catalysts for glucose oxidation by incipient wetness

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Abstract

The preparation of gold catalysts for liquid-phase glucose oxidation by the incipient wetness method was investigated. It could be shown that this preparation method is a good alternative to preparation with deposition–precipitation. TEM analysis revealed that it is possible to prepare small gold particles with particle size <2 nm by incipient wetness, even at high gold loadings. In glucose oxidation, the catalysts are very similar in terms of activity, selectivity and long-term stability to those prepared by deposition–precipitation with urea as the precipitation agent. From ecological and economical standpoints, the incipient wetness method has some advantages over the DP urea method.

Keywords: Glucose oxidation; Deposition–precipitation; DP urea; Incipient wetness; Gold catalyst

1. Introduction

Gold was long considered a less-useful noble metal for catalysis. In the 1970s, Bond [1,2] and Parravano [3] began to investigate the catalytic properties of gold and found that gold has excellent selectivity in hydrogenation and oxidation reactions, but very low activity compared with other noble metals, such as platinum and palladium. An essential breakthrough in the use of gold as a catalytic metal was the development of the deposition–precipitation (DP) and co-precipitation methods, both using sodium hydroxide as a precipitation agent, by Haruta et al. [4,5]. These authors showed that gold particles with particle sizes <4–5 nm exhibit very high activity in the oxidation of CO at and below ambient temperature, because only such small particles are able to adsorb small molecules like oxygen, hydrogen, or carbon monoxide, in contrast to larger ones.

Today, several preparation methods are used to generate catalysts with small and active gold particles [6–10]. The most frequently used preparation methods for metal oxide supports are the deposition–precipitation method, using either sodium hydroxide or urea as a precipitation agent, and the co-precipitation method. For gold on carbon catalysts, the preferred preparation method is the deposition of gold colloids. Impregnation methods, which are commonly used for the preparation of other supported metal catalysts [11,12], turn out to be not useful for the preparation of gold catalysts. In the early experiments [1–3,13,14], impregnation methods were used for gold catalyst preparation, but the particle sizes obtained were very large, resulting in very low or no catalyst activity. Later, Zanella [15] and Soares [16] reported gold particle sizes of 10 nm, with some larger particles of up to 100 nm, using the incipient wetness method. Those catalysts showed no activity in CO oxidation. Recently, several groups [17,18] described a modified incipient wetness method in which the normal impregnation step with hydrogen-tetra-chloro-aurate is followed by an additional preparation step in which the impregnated catalyst is treated with a base to achieve an exchange of chloride against hydroxyl groups. Thus, this modified incipient wetness method is more similar to the deposition–precipitation methods than to the original incipient wetness method.

Most of the knowledge about gold catalysts came from experiments concerning CO oxidation, but some of the fundamental results can be brought forward to other reactions, such as the relationship between particle size and activity. Besides use in CO oxidation, gold catalysts also can be used in other reactions, including the epoxidation of propylene [19], hydrogenation reactions such as the hydrogenation of crotonaldehyde to crotylalcohol [20], and dehydrogenation reactions such as the dehydrogenation of methanol to formaldehyde [20]. In 2002,

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Biella et al. [9] reported on the use of gold catalysts in the liquid-phase oxidation of glucose to gluconic acid. They used gold colloids immobilized on carbon and found an outstanding selectivity (100%) towards gluconic acid. This high selectivity and the significantly improved activity indicated substantial progress compared with conventional catalysts containing platinum or palladium together with lead or bismuth [21,22].

D-gluconic acid is a very interesting biodegradable product with an annual production of 100,000 t worldwide, used in the pharmaceutical, food, paper, and concrete industries [23]. The entire annual demand of gluconic acid is met via biotechnological processes involving *Aspergillus niger* and *Gluconobacter suboxydans* [23], because the bismuth- and lead-promoted platinum and palladium catalysts currently available are not suitable for technical processes. Although a high selectivity (up to 97%) can be achieved, the main drawback is the strong deactivation during the reaction, often accompanied by leaching of the second metal. The gold catalysts prepared by Biella et al. [9] showed an increased selectivity of up to 100%. However, the long-term stability of the catalysts was not sufficient; the activity decreased by about 50% within only four repeated batches. Recently, Önal et al. [24] studied similarly prepared gold colloids on carbon and their kinetics in glucose oxidation. Although they reported a lower selectivity for gluconic acid, they confirmed in principle the results of Biella et al.

Recently, we studied alumina-supported gold catalysts prepared for glucose oxidation by deposition–precipitation using either sodium hydroxide (DP NaOH) or urea (DP urea) as a precipitation agent [25]. We found that both deposition–precipitation methods led to quite active and long-term stable catalysts for glucose oxidation, but noted large differences in the suitability of the preparation methods concerning the reproducibility and the loss of gold during the preparation process. It was not possible to adjust a definite gold content using the DP NaOH method, and gold losses of up to 70% during preparation were observed. The DP urea method was found to be very suitable for the preparation of gold catalysts with very definite gold content without any loss of gold.

In this work, we prepared gold catalysts by the incipient wetness method and investigated the reproducibility of the preparation procedure and the resulting activity as well as, in particular, the long-term stability in the liquid-phase glucose oxidation. Furthermore, we compared their properties with those catalysts prepared in the previous work [25]. The preparation of supported gold catalysts by the incipient wetness method would be advantageous, because it is a very simple and widely used method that can be easily scaled up to industrial dimensions [26].

2. Experimental

2.1. Catalyst preparation

The catalysts were prepared by two different deposition–precipitation methods using either sodium hydroxide and or urea as a precipitation agent and also by the incipient wetness method. Alumina (Puralox SCFa-90, Sasol, particle size 25 µm, BET surface area 100 $m^2\,g^{-1}$, pore volume 0.5 $ml\,g^{-1}$, doped with 0.3 wt% Na_2O) was used as a support, and $HAuCl_4$ (Chempur, 50 wt% Au) was used as the gold precursor. The amount of gold on the catalysts is always given as a weight fraction (wt%). All catalysts were used for glucose oxidation directly after preparation.

2.1.1. Deposition–precipitation

2.1.1.1. Deposition–precipitation with NaOH A 1.5-g (dry mass) sample of the support was suspended in 50 ml of deionized water, and the pH was adjusted to 7 by adding HCl (0.1 $mol\,L^{-1}$) dropwise. Then 3 ml of an aqueous solution of $HAuCl_4$ (5 $g\,L^{-1}$ Au) was diluted with 50 ml of deionized water, and the pH was adjusted by adding NaOH (0.1 $mol\,L^{-1}$) dropwise. After the gold solution was poured into the support suspension, the pH was readjusted to 7. The resulting suspension was thermostatted at 80 °C for 2 h. After the suspension was cooled to room temperature, 10 ml of an aqueous solution of magnesium citrate (20 $g\,L^{-1}$) was added, and the mixture was stirred for an additional hour. Then the catalyst was treated further as described below.

2.1.1.2. Deposition–precipitation with urea In a thermostatted reactor (80 °C), 2 g of the support was suspended in 100 ml of an aqueous solution of urea (0.21 $mol\,L^{-1}$), and the specified amount of an aqueous solution of $HAuCl_4$ (5 $g\,L^{-1}$ Au) was added quickly. The suspension was vigorously stirred for 20 h and then cooled to room temperature.

2.1.1.3. Further treatment After the gold precursor was deposited onto the support, all catalysts were treated in the same way: (i) separation from the preparation suspension by filtering through a sintered-glass filter (porosity 4); (ii) washing with 200 ml of deionized water to remove residual Cl^- ions and Au species that were not deposited on the support; (iii) drying overnight at 80 °C; and (iv) calcination in air at 200 °C.

2.1.2. Incipient wetness

The required amount of $HAuCl_4$ was dissolved in a volume of deionized water corresponding to the pore volume of the support. This solution was always freshly prepared and was added dropwise to the support during intensive mixing. At the completion of the addition, the support was slightly wet. The addition of the impregnation solution took about 15 min. Immediately afterwards, the resulting precursor was dried for 16 h at 80 °C and subsequently reduced in the gas phase using 5 vol% hydrogen in nitrogen at 250 °C for 2 h.

2.2. Catalyst characterization

The gold content of the prepared catalysts was determined by inductively coupled plasma–atom emission spectrometry (ICP-AES), using an Integra XM system (GBC). The catalysts were digested with a mixture of 1 ml HNO_3 (65%), 3 ml HCl (37%), and 5 ml HF (40%) in a microwave.

Some of the catalysts were investigated by temperature-programmed reduction (TPR) and TEM. The TPR measurements were done by heating the catalyst precursor in a gas flow of 5 vol% hydrogen in argon at a heating rate of 10 $\mathrm{K\,min^{-1}}$ from room temperature up to 320 °C in a TPR/TPD 2900 system (Micromeritics).

Particle sizes were determined by TEM analysis using a CM20 TWIN (LaB6) operating at 200 kV at the Leibniz Institute of Catalysis at the University of Rostock, Berlin Branch.

2.3. Catalytic testing

All catalysts were tested in the aqueous phase glucose oxidation as a model reaction. The reactions were carried out at 40 °C in a thermostatted glass reactor (500 ml). To avoid mass transfer limitations, the amount of catalyst was adjusted between 0.01 and 0.10 g, depending on the gold content. During the experiment, the pH of the reaction suspension was kept constant at 9 by the addition of 2.5 $\mathrm{mol\,L^{-1}}$ NaOH using a Titroline alpha system (Schott). Oxygen was bubbled through the suspension with a flow rate of 500 $\mathrm{ml\,min^{-1}}$ at atmospheric pressure. The suspension was stirred with a magnetic stirrer at 700 rpm. The reactions were carried out until 100% conversion was reached. At the end of each reaction, conversion and selectivity were checked by HPLC using a Luna Amino 5 μ column (Phenomenex). Because the selectivity was 100% in all cases, it was possible to calculate the activity directly from the titration curve of the resulting gluconate. ICP-AES analyses revealed no aluminium leaching at the reaction conditions.

The specific activity, related to the gold content of the catalysts, was used to compare catalysts with different gold contents. Some catalysts showed an induction period, so the maximum activity was calculated for comparison as outlined previously [25].

The catalyst's long-term stability was investigated by repeated batches of glucose oxidation. The catalyst was separated by filtration after complete conversion after one batch, washed with water, and dried overnight at 70 °C. Subsequently, it was used in the next batch. Before the first batch and after the last batch, the gold content of the catalyst was determined by ICP-AES to check whether any gold leaching occurred.

3. Results and discussion

3.1. Catalyst preparation

Recently, we reported on the preparation of gold catalysts for the glucose oxidation by deposition–precipitation using sodium hydroxide and urea as precipitation agents. We found that both DP methods led to highly active and long-term stable catalysts [25]. But significant differences existed in terms of the mechanisms occurring during preparation. When sodium hydroxide was used as a precipitation agent, the pH of the precipitation solution had two opposing effects. Due to the better electrostatic interactions at lower pH (e.g., pH 5–6) nearly all of the gold could be deposited on the support. But the gold particles resulting from the gold complexes present at this pH were too large to be active in catalytic reactions. Increasing the pH led to smaller and thus more active gold particles on the support but, due to lower electrostatic interactions, a smaller amount of gold that could be deposited on the support. Thus, during the preparation with sodium hydroxide, gold loss of up to 70% was observed. When urea was used as a precipitation agent, no real deposition–precipitation mechanism occurred; rather, an impregnation mechanism was seen, followed by a ligand exchange of the adsorbed gold complexes in which chloride was exchanged against hydroxyl ions. With urea as a precipitation agent, all of the gold offered could be deposited on the precursor over the entire range of gold loadings (up to 10 wt% gold) investigated.

Table 1
Gold contents of catalysts prepared by incipient wetness

Catalyst	Theoretical Au content (wt%)	Experimental Au content (wt%)
IW01	0.30	0.32
IW02	1.00	1.01
IW03	3.00	3.06
IW04	5.00	4.95

The complete deposition of the intended amount of gold on the support is a prerequisite for a suitable preparation method. To evaluate this deposition for the incipient wetness method described in the present paper, four catalysts with gold loadings in a range of 0.3 up to 5 wt% were prepared and analyzed by ICP-AES. The results are shown in Table 1.

No loss of gold was observed during the preparation procedure. This finding can be understood by examining the mechanism of the incipient wetness method. Deposition of the active compound was achieved by filling the impregnation solution directly into the pores of the support. The volume of this impregnation solution corresponds to the empirically determined pore volume of the support. At the completion of the addition, the support began to look wet. The impregnation step was followed by a drying step and subsequent gas-phase reduction. There was no washing step or any other step during the incipient wetness preparation procedure in which a loss of gold could occur.

In addition, there should be a strong electrostatic interaction between gold complexes in the impregnation solution and the support. The pH of the impregnation solution is acidic, because hydrogen-tetra-chloro-aurate is an acid. Because hydrogen-tetra-chloro-aurate is not stable in water, it begins to hydrolyse in the impregnation solution. Due to this hydrolysis, the gold in the impregnation solution is present in the form of anionic complexes containing chloride and hydroxyl ions (e.g., $[\mathrm{AuCl_3(OH)}]^-$, $[\mathrm{AuCl_2(OH)_2}]^-$). Due to the acidic pH of the impregnation solution, the surface of the alumina support is protonated and positively charged. Thus, the negatively charged chloride-containing gold complexes should strongly adsorb on the positively charged support surface.

These chloride-containing gold complexes seem to be responsible for the failure of the incipient wetness method in the past. Gold catalysts prepared by the incipient wetness method have always been found to be unsuitable because the resulting gold particles are quite large even at low gold content. Several authors [20,27–29] have shown that chloride enhances the mobility and agglomeration of gold species during the calcination

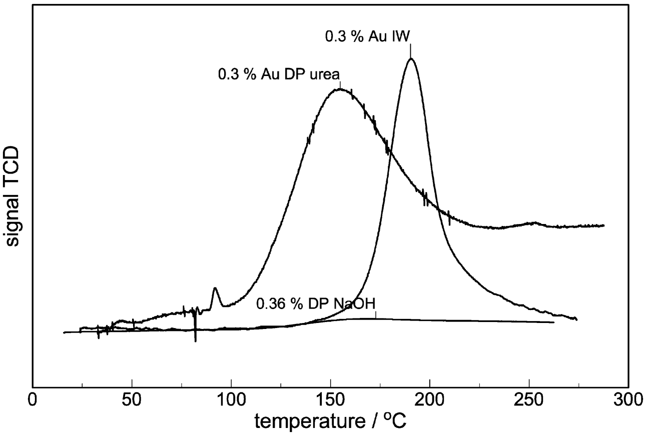


Fig. 1. Temperature-programmed reduction (TPR) of the gold catalysts prepared by DP NaOH (0.36% Au), DP urea (0.3%) and incipient-wetness (0.3% Au).

process. We decided to use a gas-phase reduction procedure with hydrogen instead of the calcination procedure, to avoid agglomeration of the gold species during the calcination process. During the gas-phase reduction with hydrogen, chloride likely is discharged as hydrogen chloride by the gas flow.

The different mechanisms of the deposition–precipitation and incipient wetness methods lead to the conclusion that different types of gold species should be present on the nonreduced catalyst precursors. To determine whether different gold species exist on the precursors after deposition of the active compound, we investigated the noncalcined and nonreduced catalyst precursors of each preparation method by TPR. Significantly different reduction temperatures are a clear indication for different metal species on an identical support or for different interactions of the same species with different supports. Because in this case the support is the same in all cases, different reduction temperatures are an indication for different gold species. Fig. 1 shows TPR profiles for a 0.30 wt% Au/Al_2O_3 catalyst prepared by DP urea, a 0.36 wt% Au/Al_2O_3 catalyst prepared by DP NaOH, and a 0.30 wt% Au/Al_2O_3 catalyst prepared by incipient wetness. The three curves are quite different. It is striking that the curve for the DP NaOH precursor is very flat, indicating low hydrogen consumption by this catalyst. The reason for this low hydrogen consumption might be that the catalyst already was reduced during the preparation process. This reduction may have resulted from the use of magnesium citrate, which can act as a reducing agent for gold compounds. The maxima of the three curves indicating the reduction temperatures of the catalysts are quite different as well. The two catalysts prepared by deposition–precipitation using sodium hydroxide or urea as precipitation agents have quite similar reduction temperatures (160 °C DP urea; 170 °C DP NaOH), whereas the reduction temperature for the incipient wetness catalyst is significantly higher at 190 °C. The similar reduction temperatures of the two DP catalysts are an indication of quite similar gold species on the precursors.

In our previous study [25], we surmised that the gold complexes present on the precursors prepared by the deposition–precipitation method contained only small amounts of chloride. Due to the nearly complete hydrolysis of the tetra-chloro-aurate ion, most of the chloride should be substituted by hydroxyl ions. When urea is used as a precipitation agent, the complexes likely also contain some ammonia ligands. The significantly higher reduction temperature of the incipient wetness catalyst is an indication of a quite different gold species on this precursor. Because of the hydrolysis chemistry of the tetra-chloro-aurate anion, it is verisimilar that the gold complex present on this precursor is an anionic complex containing large amounts of chloride ($[AuCl_3(OH)]^-$), as already outlined.

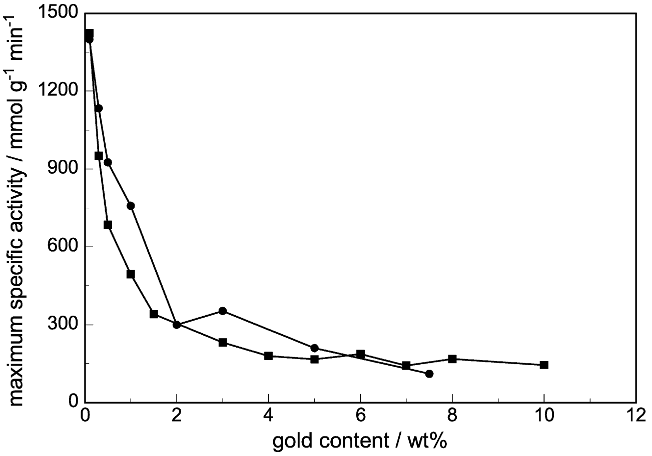
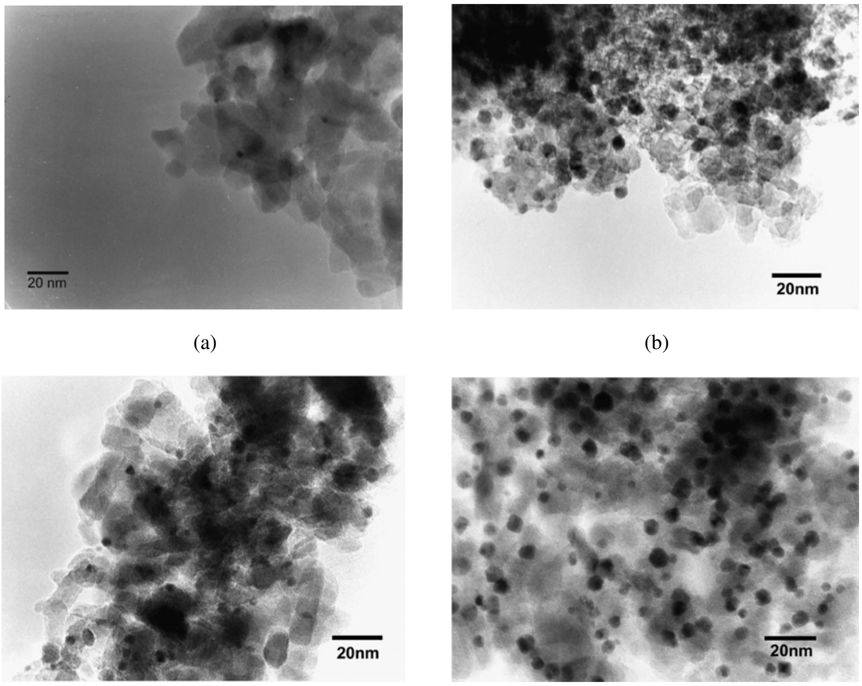


Fig. 2. Dependency of the maximum specific activity on the gold content of catalyst prepared by DP urea (■) and incipient-wetness (●).

3.2. Dependence of particle size and activity on gold content

Our previous investigation of catalysts prepared by DP urea showed a strong dependence of specific activity on the gold content [25]. The investigation of the activities of the gold catalysts prepared by incipient wetness found a quite similar relationship. Fig. 2 shows the curves of the dependence of specific activity on the gold content for gold loadings ranging from 0.1 to 10 wt% for the DP urea method and from 0.1 to 7.5 wt% for the incipient wetness method. Very high activity was observed at very low gold content. Increasing the gold content led to a decrease in catalyst activity. From gold contents of about 4 wt% to higher gold amounts, the specific activity remained constant. It is noteworthy that the activities of catalysts prepared by the two preparation methods are very similar over the entire range investigated.

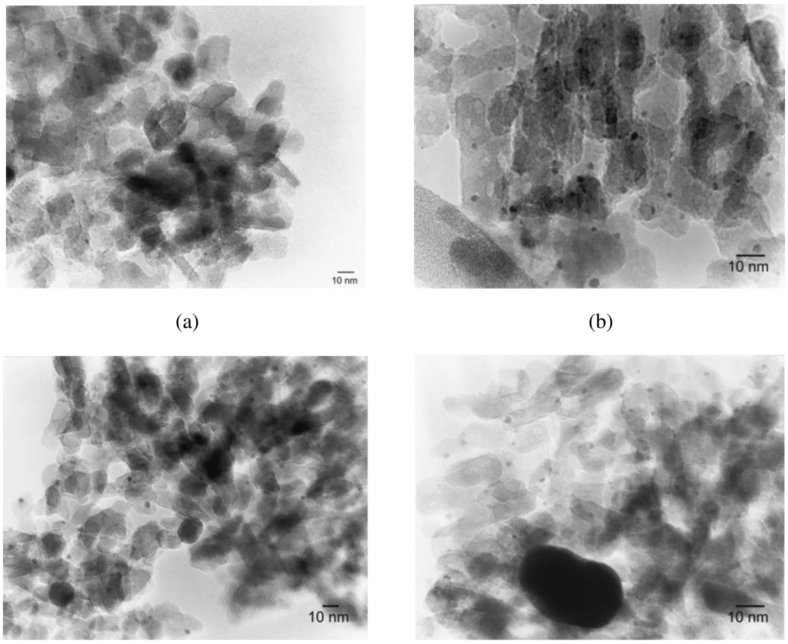
Analysis of the gold particle size of selected catalysts by TEM should help determine whether a correlation exists among gold content, activity, and particle size. TEM images of several selected catalysts with various gold contents prepared by both preparation methods are shown in Figs. 3 (DP urea) and 4 (incipient wetness). The TEM images of highly active catalysts with low gold content (0.1 wt% [DP urea] and 0.3 wt% [incipient wetness]) show extremely small gold particles (1.2–3 nm) for both catalysts. The average particle size of the catalyst with the lower gold content (0.1 wt% DP urea) is somewhat smaller (1.6 nm) than for the catalyst with 0.3 wt% (incipient wetness), which has an average particle size of 1.9 nm. This difference



(a) (b)

(c) (d)

Fig. 3. TEM analysis of catalysts prepared by DP urea: (a) 0.1 wt% Au; (b) 0.5 wt% Au; (c) 1.5 wt% Au; (d) 10 wt% Au.



(a) (b)

(c) (d)

Fig. 4. TEM analysis of catalysts prepared by incipient-wetness: (a) 0.3 wt% Au; (b) 1 wt% Au; (c) 3 wt% Au; (d) 5 wt% Au.

in average particle size is in good agreement with the different activities of these catalysts. The catalyst with 0.3 wt% gold content has a slightly lower activity (1100 $\text{mmol g}^{-1}\,\text{min}^{-1}$) than the catalyst with 0.1 wt% gold. An increase in the gold content leads to an increase in particle size for both preparation methods. For the catalyst with 1.0 wt% gold content prepared by incipient wetness, we found gold particle sizes of 1.5–6 nm, with an average particle size of 2.8 nm. Further increases in gold content up to 3 and 5 wt% (incipient wetness) led to catalysts with three different sizes of gold particles: very small (2–5 nm) particles, a few medium-sized particles (10–25 nm), and sporadic large particles (ca. 50 nm for the 3 wt% catalyst and up to 200 nm for the 5 wt% catalyst). The catalyst with 10 wt% gold prepared by DP urea exhibited two types of gold particles: small particles (2–10 nm) and some quite large particles (up to 30 nm).

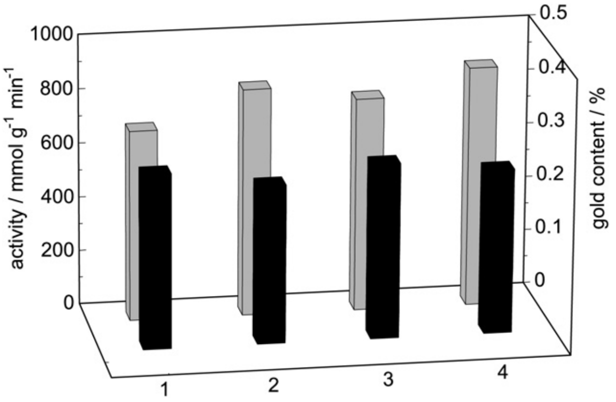


Fig. 5. Reproducibility of activity (▨) and gold content (■) of four 0.3 wt% gold catalysts prepared by incipient-wetness.

A strong dependence of catalytic activity on gold particle size has been described mainly for gas-phase reactions [6,30]. For liquid-phase reactions, Comotti et al. [31] investigated the relationship between the size of gold colloids and their activity in glucose oxidation and found that particles must be smaller than at least 10 nm to be active in glucose oxidation. As mentioned earlier in this paper, such small gold particles are present on every catalyst, even on those with high gold loadings. Thus, we can for the first time provide evidence that preparing gold catalysts with gold particles smaller than 10 nm is possible by the incipient wetness method, even at gold loadings up to 5 wt%. In 2006, Hutchings et al. [32] reported on gold catalysts prepared on carbon by an incipient wetness method quite similar to that used in the present work. Those catalysts were used in the oxidation of glycerol, cyclohexane, and primary alcohols but were not active in CO oxidation. Unfortunately, these authors provided no information on the gold particle sizes that they achieved using this preparation method.

Although numerous small gold particles are present on the catalysts with high gold loadings prepared by either of the two preparation methods, a significant part of the gold is located in catalytically nonactive particles larger than 10 nm. Taking into account only those gold particles smaller than 10 nm (i.e., those active for glucose oxidation according to the findings of Comotti et al. [31]), the TEM micrographs in Figs. 3d and 4d reveal an average size of those small particles of 4.5 nm for the 10 wt% Au catalyst prepared by DP urea and 3.1 nm for the 5 wt% Au catalyst prepared by the incipient wetness method. On the basis of these average sizes, a turnover frequency can be estimated according to the magic cluster model [33] by assuming that every surface atom is an active site. This estimation leads to the conclusion that the 5 wt% Au catalyst prepared by incipient wetness should be 1.3 times more active than the 10 wt% Au catalyst prepared by DP urea, which is in good agreement with the experimentally determined value of 1.45.

3.3. Reproducibility of the incipient wetness method

The application of the gold catalysts prepared by the incipient wetness method showed that these catalysts are as active in glucose oxidation as catalysts prepared by DP methods. HPLC analysis revealed a selectivity toward sodium gluconate of 100%, just as for the DP catalysts. In our previous work [25], we showed that the DP urea method not only leads to active and highly selective catalysts, but also is a highly reproducible preparation method in terms of both activity and gold content. In contrast, the DP NaOH method was not reproducible in terms of the gold content or the resulting activity.

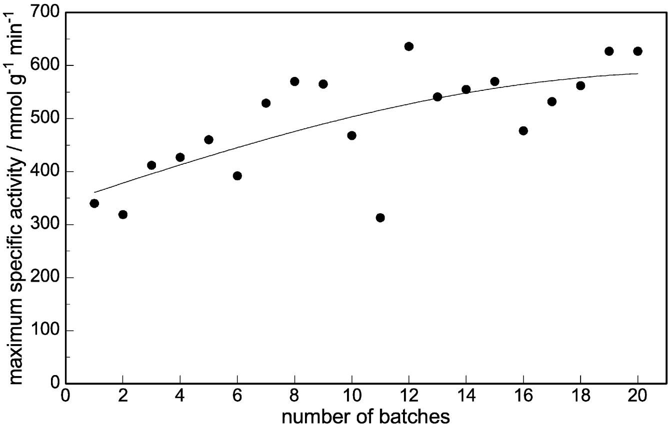


Fig. 6. Long-term stability of a 0.3 wt% gold catalyst prepared by incipient-wetness in glucose oxidation.

Because the reproducibility of the catalyst properties is essential with regard to their application in a technical process, we attempted to determine the reproducibility of the incipient wetness method. We prepared four individual catalyst charges under identical conditions, each with an intended gold content of 0.3 wt%. The resulting gold content was checked by ICP-AES, and the activity was tested in glucose oxidation. The results, given in Fig. 5, show that the gold content is as reproducible as the activity.

3.4. Catalyst durability

Based on the results obtained so far, incipient wetness appears to be a very promising method of preparing gold catalysts for liquid-phase oxidations. We determined the long-term stability of a 0.3 wt% gold catalyst prepared by the incipient wetness method. We used the catalyst in 20 repeated batches of glucose oxidation (Fig. 6) and found an increase in activity in the first 8 batches. In batches 9–20, the activity remained fairly constant, with some fluctuations. The increased activity in the first 8 batches was caused not by the catalyst itself, but rather by a diminishing oxygen mass transfer limitation. Because a loss of catalyst mass occurs after each batch during the filtration procedure, it was necessary to start the repeated batch experiment with such a high amount of catalyst that oxygen mass transfer limitations occur in the beginning. After the eighth batch, the amount of catalyst decreased to an extent that no more limitations remained. The selectivity towards sodium gluconate was 100% in all batches. ICP-AES analysis before the first batch and after the last batch showed no loss of gold during the experiment. These findings reveal that the catalysts prepared by the incipient wetness method have excellent long-term stability.

The results of the present work demonstrate that the incipient wetness method is a promising alternative method of preparing gold catalysts for liquid-phase oxidation. In concert with the DP method using urea as a precipitation agent, two methods of preparing gold catalysts that are active, selective and stable in liquid-phase glucose oxidation are now available. Both preparation methods fulfil the requirement that all of the gold is deposited on the catalyst during the preparation process. However, the DP urea method has the drawback of producing large amounts of wastewater; preparation of 2 g of catalyst produces about 500 ml of ammoniacal wastewater. Wastewater treatment is an expensive additional process step that must be taken into account in an industrial process. In contrast, the incipient wetness preparation procedure produces no wastewater. A slight drawback of the incipient wetness method is for safety reasons the need to use hydrogen for catalyst reduction. However, on an industrial scale, the gas-phase reduction of catalysts is a common process step; thus, the incipient wetness method seems more suitable for the production of gold catalysts on an industrial scale, as described by Corti et al. [26].

4. Conclusion

This paper has demonstrated that the incipient wetness method is an excellent alternative method of preparing gold catalysts for liquid-phase glucose oxidation. Catalysts prepared by this method have activities comparable with those prepared by the DP urea method. The incipient wetness catalysts are 100% selective toward sodium gluconate and have sufficient long-term stability. TEM demonstrated an increased particle size with increasing gold content for both the DP urea and incipient wetness methods. In addition, we found that it is possible to prepare gold catalysts containing small gold particles with the incipient wetness method, even at high gold content. From ecologic and economic standpoints, the DP urea and incipient wetness methods should be favoured, because these are the only methods available for preparing gold catalysts with specific gold contents and no gold loss. The incipient wetness method should be better suited than the DP urea method for producing gold catalysts on an industrial scale, because it produces no wastewater.

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