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New innovative gold catalysts prepared by an improved incipient wetness method

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ABSTRACT

We described 0.3% Au/Al_2O_3 catalysts prepared by the incipient wetness impregnation method using hydrogen tetrachloroaurate dissolved in different aqueous solutions as an impregnation solution. Hydrogen tetrachloroaurate hydrolyses in aqueous solution and the composition of the gold complexes depend on the pH value and the chloride concentration; therefore, different impregnation solutions were prepared by dissolving the hydrogen tetrachloroaurate in water and aqueous solutions of hydrochloric acid, nitric acid, sodium chloride, and sodium hydroxide. The resulting gold complexes were evaluated by UV/vis spectroscopy. Gold catalyst precursors were activated via gas-phase reduction with hydrogen. In addition, the catalyst precursors and the catalysts were investigated by TPR and TEM analysis, respectively, and tested in glucose oxidation. The gold catalyst prepared with a hydrochloric acid solution exhibited the highest activity in glucose oxidation and the smallest gold particles (mean diameter, 1–2 nm). In fact, the catalysts prepared from precursor solutions in which the tetrachloroaurate ion was stabilized demonstrated the highest activities. This is a significant finding, because residual chloride in the catalyst precursor has been considered detrimental to the preparation of active catalysts. In addition, the gold catalyst prepared with 2 M HCl was tested in repeated batches and exhibited excellent long-term stability for the conversion of highly concentrated glucose solutions.

1. Introduction

Gold catalysis has attracted increasing attention over last two decades. Although gold supported on metal oxides has been known to be highly selective in hydrogenation and oxidation processes since the 1970s, their low activity has discouraged broader applications [1–3]. Later, it was found that the activity of gold catalysts depends strongly on the presence of small gold particles. The first suitable preparation methods for supported nanoscale gold catalysts, deposition–precipitation (DP) and co-precipitation methods using sodium hydroxide as the precipitation agent, were not developed until the late 1980s [4,5]. These catalysts were used for low-temperature CO oxidation and demonstrated high activity only when gold nanoparticles smaller than 5 nm were present.

Today, several preparation methods are known to produce active gold catalysts [6–8]. The most common ones are the DP of gold on metal oxide supports and the immobilization of gold colloids on carbon. Impregnation methods are commonly used for the preparation of other supported metal catalysts, because they are simple and easy to scale up. But these methods have long been considered unsuitable for gold catalysts, because the prepared catalysts had gold particles larger than 5 nm [9,10] and demonstrated very little activity (in, e.g., CO oxidation) [9].

Recently, modified impregnation methods have been reported in which, after the support is impregnated with hydrogen tetrachloroaurate, the catalyst is treated in an additional step with base to achieve an ion exchange of chloride against hydroxyl [11,12]. Residual chloride ligands were held to be responsible for the low activity of gold catalysts in previous studies [9,10,13–15], due to gold particle agglomeration observed during the calcination of chloride-containing gold precursors, as well as a poisoning effect of chloride on active gold itself for CO oxidation [14]. Nevertheless, we recently showed that it is possible to produce gold catalysts with small gold particles (1.5–6 nm) using an incipient wetness impregnation method with hydrogen tetrachloroaurate with no exchange of chloride after preparation [16]. These catalysts were activated not by calcination, but rather by gas-phase reduction with hydrogen. The reduction with hydrogen is a simple, industrially feasible method for the production of gold catalysts. These catalysts demonstrated very high activity, selectivity, and also long-term stability in the liquid-phase glucose oxidation toward gluconic acid.

Hydrogen tetrachloroaurate is the main gold precursor used for catalyst preparation by DP and impregnation methods. In aqueous media, hydrogen tetrachloroaurate(III) hydrolyses, resulting various species with different amounts of chloride, water, and hydroxyl (e.g., $[AuCl_4]^-$, $[AuCl_3(H_2O)]$, $[AuCl_3OH]^-$, $[AuCl_2(OH)_2]^-$, $[AuCl(OH)_3]^-$, $[Au(OH)_4]^-$); their composition depends on the pH value and the chloride concentration [17]. For DP methods, the ac-

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tivity of a gold catalyst for CO oxidation is known to increase with increasing hydrolysis of the tetrachloroaurate ion if the precursors are activated by calcinations [18]. Therefore, we investigated the influence of differently hydrolysed gold complexes in the impregnation solution on the activity of gold catalysts prepared by incipient wetness. Different impregnation solutions were prepared by dissolving hydrogen tetrachloroaurate in aqueous solutions with different pH's and/or different chloride concentrations. The natures of the different gold complexes were determined by UV/vis spectroscopy, and the catalysts were investigated by TPR and TEM analysis. The activity of all catalysts was tested in liquid-phase glucose oxidation. In addition, the long-term stability of the most active catalyst was tested in repeated batches with high, industrially relevant glucose concentrations.

2. Experimental

2.1. Catalyst preparation

Alumina (Puralox KR-90, Sasol; particle size 25 µm, BET surface area 90 $m^2\,g^{-1}$, pore volume 0.6 $ml\,g^{-1}$) was used as the support. $HAuCl_4$ (Chempur, 50 wt% Au) was used as the gold precursor. All catalysts were tested in glucose oxidation immediately after preparation.

The catalysts were prepared by the incipient wetness method and contained 0.3 wt% gold. The required amount of solid $HAuCl_4$ (notation $HAuCl_{4(s)}$) was dissolved in a volume of varying aqueous media, each corresponding to the pore volume of the support. The following aqueous media were used: 2 M HCl, 2 M NaCl, 2 M HNO_3, 0.25 M NaOH, and pure water. After dissolving the $HAuCl_4$ in the aqueous media, the resulting pH values of the impregnation solutions were <0 for 2 M HCl and 2 M HNO_3, $\sim$1.5 for 2 M NaCl and water, >13 for 0.25 M NaOH. The impregnation solution was added dropwise to the support during intensive mixing. After addition of the solution was complete, the support became slightly wet. The resulting catalyst 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.

In one case, a stock solution of 25 $g\,l^{-1}$ $HAuCl_4$ that had been aged for several days was used instead of solid $HAuCl_4$, and the amount of water needed to reach the required volume of impregnation solution (notation $HAuCl_{4(aq)}$) was added. The pH value of this solution was $\sim$1.5. For comparison, another 0.3% Au/Al_2O_3 catalyst was prepared by the deposition precipitation method with NaOH according to the procedure described in [27].

2.2. Catalytic testing

All catalysts were tested in aqueous-phase glucose oxidation as a model reaction. The reactions were carried out at 40 °C in a thermostatted glass reactor with a reaction volume of 500 ml. The glucose concentration was 100 $mmol\,l^{-1}$. During the experiment, the pH of the reaction suspension was kept constant at 9 through addition of 2.5 $mol\,l^{-1}$ NaOH. Oxygen was bubbled through the suspension with a flow rate of 500 $ml\,min^{-1}$ 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. Because the selectivity was $>99\%$ in all cases, it was possible to calculate the activity directly from the titration curve of the resulting gluconate. The specific activity was related to the gold content.

The catalyst's long-term stability was investigated at the much higher glucose concentration of 30 wt%, which would be a reasonable concentration in a potential industrial process. During the repeated batches 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.

2.3. Analytical methods

UV/vis spectra of the different impregnation solutions were measured using a Specord 40 (Analytikjena). The gold content of the prepared catalysts was determined by inductively coupled plasma-atom emission spectroscopy (ICP-AES), using an Integra XM system (GBC). Therefore, 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) in a Micromeritics TPR/TPD 2900 system. The TPR measurements were carried out by heating the catalyst precursor with a rate of 10 $K\,min^{-1}$ from room temperature up to 350 °C in a H_2/Ar gas flow (5 vol% H_2). A sodium hydroxide trap was used as gaseous acids could be released during the TPR procedure.

Gold 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 Rostock, Branch Berlin. Conversion, selectivity, and mass balance of each reaction were checked by HPLC using a Luna Amino 5 µm column (Phenomenex) and 20 $mmol\,l^{-1}$ H_3PO_4 as a mobile phase.

3. Results and discussion

3.1. UV/vis spectra of impregnation solutions with different pH and chloride concentration

In recent work [16], we prepared gold catalysts for glucose oxidation were prepared by a classical incipient wetness preparation method using gas-phase reduction with hydrogen. These catalysts were found to be as active and selective in glucose oxidation as catalysts prepared by DP methods. During the incipient wetness preparation, solid hydrogen tetrachloroaurate was dissolved in a volume of deionized water corresponding to the pore volume of the used support. The pH of the impregnation solution was acidic, because hydrogen tetrachloroaurate is a strong acid that begins to hydrolyse in aqueous solution. Due to this hydrolysis, the gold in the impregnation solution is present in the form of anionic complexes containing chloride and hydroxyl ions [17]. The hydrolysis of the tetrachloroaurate complex can be described as

$$[AuCl_{4-i}OH_i]^- + H_2O = [AuCl_{3-i}OH_iH_2O] + Cl^-,$$

$$[AuCl_{3-i}OH_iH_2O] = [AuCl_{3-i}OH_{i+1}]^- + H^+ \quad (i = 0\text{–}3).$$

During the hydrolysis, chloride is substituted stepwise against hydroxyl. The composition of the different gold complexes in the condition of equilibrium depends mainly on the concentration of chloride ions and the pH.

The dependence of the hydrolysis of hydrogen tetrachloroaurate at different pH values was described by Peck et al. [19]. They used UV–vis spectroscopy to show that at constant chloride concentration the tetrachloroaurate ion is favoured with decreasing pH. Murphy et al. [20] and Farges et al. [21] described that the hydrolysis of the tetrachloroaurate ion is lower in the presence of excess chloride, even at higher pH values. Ivanova et al. [22] also detected the tetrachloroaurate ion at low pH values using UV/vis spectroscopy. But because they used hydrochloric acid to adjust the low pH values, they changed not only the pH value, but also

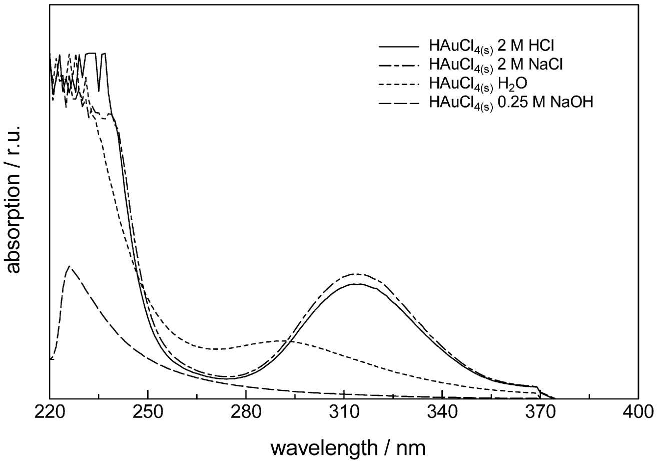


Fig. 1. UV–vis spectra of different hydrogen tetrachloroaurate solutions.

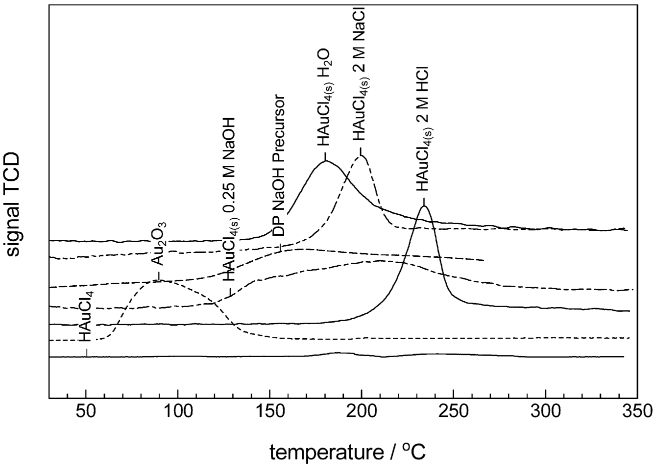


Fig. 2. TPR profiles of different gold precursors prepared by incipient wetness method.

the chloride concentration. Therefore, parts of the tetrachloroaurate could have been stabilized by the additional chloride ions. To differentiate between the effect of pH and chloride, different hydrogen tetrachloroaurate solutions were prepared using solutions with different pH and chloride concentrations. Low pH was achieved by adding 2 M nitric acid and high pH by adding 0.25 M sodium hydroxide solution. As no additional chloride was used, in this case, the effects on the chloroaurate complexes were related to the pH. To obtain high chloride concentrations without changing the pH, 2 M sodium chloride solution was added. High chloride concentration and low pH were adjusted using 2 M hydrochloric acid. In addition, an impregnation solution was prepared with water as described previously [16]. UV/vis spectroscopic analysis of the impregnation solution should give information about the hydrolysis of the tetrachloroaurate ion [19]. Fig. 1 shows the UV/vis spectra obtained for selected catalysts.

The impregnation solution of the $HAuCl_{4(s)}$ 2 M HCl catalyst showed an intense band at 313 nm. This band can be ascribed to a ligand-to-metal charge transfer transition in the tetrachloroaurate ion $[AuCl_4]^-$ [19]. The same band was seen for the $HAuCl_{4(s)}$ 2 M NaCl impregnation solution. Although the hydrochloric acid solution was much more acidic, in both impregnation solutions the tetrachloroaurate ion was the dominant species. Thus, high chloride concentrations stabilized the tetrachloroaurate ion as well as HCl. In contrast, the hydrogen tetrachloroaurate dissolved in water ($HAuCl_{4(s)}$ H_2O) showed a band blue-shifted to 291 nm with diminished intensity. This indicates an initial hydrolysis of the tetrachloroaurate ion, because a blue shift is caused by the exchange of chloride for hydroxyl ions [19]. The UV/vis spectrum of the impregnation solution of the $HAuCl_{4(s)}$ 0.25 M NaOH catalyst showed no band in the area investigated, indicating complete hydrolysis of the tetrachloroaurate ion. No UV/vis spectrum could be measured for the $HAuCl_{4(s)}$ HNO_3 solution, because the NO_3^--ion interfered in the relevant wavelength section. When water was used as the solvent, the tetrachloroaurate ion started to hydrolyse, and mixed hydroxychloroaurate complexes were formed.

3.2. TPR profiles

As shown by the UV–vis spectra, the gold complexes differed in the different impregnation solutions. To check the properties of the resulting gold precursor after drying, TPR profiles were analysed. These TPR profiles are shown in Fig. 2. The precursor of the $HAuCl_{4(s)}$ 2 M HNO_3 catalyst could not be analyzed, because the thermal decomposition of HNO_3 interfered with the TCD signal of the TPR analyzer.

The reduction, and thus the reduction temperature, are related to the redox potential; the lower the redox potential, the more energy-consuming the reduction. The redox potential of gold in aqueous solutions depends on the coordinating ligands [23]. Chloride ions cause a decrease in the redox potential compared with H_2O/OH^- ligands; thus, chloride-containing gold complexes should exhibit the highest reduction temperature. The measured reduction temperatures showed the expected behaviour. The precursor prepared with hydrochloric acid ($HAuCl_{4(s)}$ 2 M HCl) exhibited the highest reduction temperature, in a quite sharp peak, at 234 °C; the $HAuCl_{4(s)}$ 2 M NaCl precursor exhibited a lower reduction temperature, at 200 °C. Although the same gold complex was present in both impregnation solutions, their reduction temperatures were markedly different. This difference likely originated from the counter-ions of chloride (i.e., H^+ and Na^+), which were still present in the dried catalyst precursor. Obviously, reduction of the tetrachloroaurate complex in an acidic environment (protonated surface hydroxyl groups) required a higher temperature than reduction of the same complex in a less acidic environment. The next-lower reduction temperature was obtained by the $HAuCl_{4(s)}$ H_2O precursor. As shown by the UV–vis investigations, the hydrogen tetrachloroaurate hydrolysed in water. In this case, chloride was partly substituted for hydroxyl in the impregnation solution, which increased the redox potential and decreased the reduction temperature. The precursor prepared with sodium hydroxide solution ($HAuCl_{4(s)}$ 0.25 M NaOH) showed a rather broad peak already at low temperature. The UV–vis spectra of the impregnation solution exhibited no tetrachloroaurate. Indeed, the reduction peak was similar to that of a precursor prepared by DP with sodium hydroxide (DP NaOH), in which chloride was exchanged against hydroxyl during preparation.

For comparison, pure hydrogen tetrachloroaurate and gold oxide also were reduced by TPR. The gold oxide had a low reduction temperature, with a maximum at 89 °C. Hydrogen tetrachloroaurate showed only negligible hydrogen uptake, likely due to the formation and decomposition of gold chloride. It likely decomposed to gold, chlorine, and hydrogen chloride, so that no hydrogen was needed for reduction and no TPR signal could be obtained. In contrast, all gold precursors exhibited hydrogen uptake, even those in which the tetrachloroaurate ion was stabilized. Like the pure hydrogen tetrachloroaurate, these precursors contained the tetrachloroaurate complex. But because they demonstrated no decomposition behaviour, like the pure hydrogen tetrachloroaurate, a strong interaction between the tetrachloroaurate complex and the alumina support was highly likely.

Nechaev et al. described the adsorption of hydrogen tetrachloroaurate on alumina [17] and found that adsorption depends on the pH and the chloride concentration. At acidic pH, the surface of the alumina support is protonated and positively charged, because of its isoelectric point of 7–9. Thus, the negatively charged chloride-containing gold complexes are strongly adsorbed on the positively charged support surface, resulting in a strong interaction. Thus, good dispersion can be reached. But changing the pH value and/or the chloride concentration changes the amount of ions in the solution as well. Because each dissolved species has a specific adsorption activity with regard to the support, numerous complex interactions occur in the solution/support mixture. The extent to which the adsorption of the gold species occurs on the support depends mainly on two factors: the ion–support ratio and the intensity of ion–support interactions.

In cases with greater amounts of additional ions (e.g., Cl^-) able to competitively adsorb on the support, the probability of adsorption of the negatively charged gold complexes declines. Very high quantities of additional ions may even lead to suppression of gold adsorption. Ivanova et al. [22] reported that they could not detect any adsorption of gold ions on alumina during their wet impregnation experiments when they used hydrochloric acid solution as the solvent. The volume of the impregnation solution is much lower in incipient wetness impregnation than in wet impregnation. Accordingly, the concentration of the gold complex is much higher in the incipient wetness impregnation solution than in the wet impregnation solution at the same hydrochloric acid concentration. Thus, in incipient wetness impregnation, the adsorption of Au species on alumina support is considerably more likely because of less competitive adsorption of other ions present.

Consequently, to obtain active catalysts, strong interactions between metal and support must be generated while maintaining a reasonable level of competitive adsorption. The incipient wetness preparation method is well suited for this purpose, because it requires only a small volume of impregnation solution.

3.3. Glucose oxidation

Recently, we demonstrated that gold catalyst prepared by incipient wetness has similar activity to gold catalysts prepared by DP [16]. Those incipient wetness catalysts were prepared with hydrogen tetrachloroaurate dissolved in water as an impregnation solution. In the present study, we investigated the influence of gold catalysts from different impregnation solutions containing different gold complexes on their activity in glucose oxidation. The specific activities of the catalysts are shown in Fig. 3. The catalyst prepared with water ($HAuCl_{4(s)}$ H_2O) exhibited similar activity as the corresponding catalyst in our previous work [16]. Both catalysts were freshly prepared by dissolving solid hydrogen tetrachloroaurate in water and using the resulting solution as an impregnation solution. In contrast, a catalyst prepared from a stocked hydrogen tetrachloroaurate solution ($HAuCl_{4(aq)}$ H_2O) exhibited lower activity. This effect may be due to the fact that the hydrolysis equilibrium of hydrogen tetrachloroaurate in water requires up to 24 h [24].

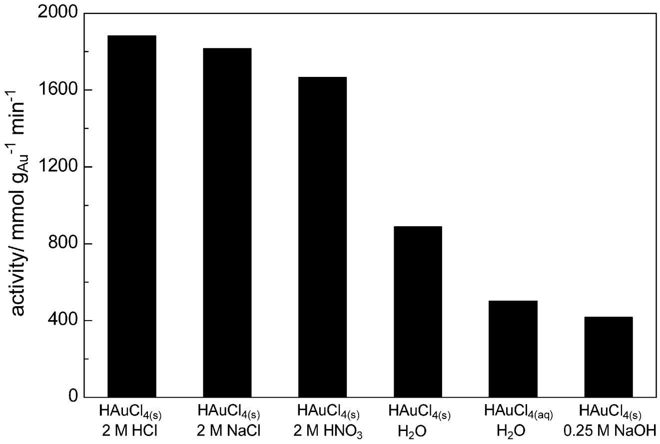
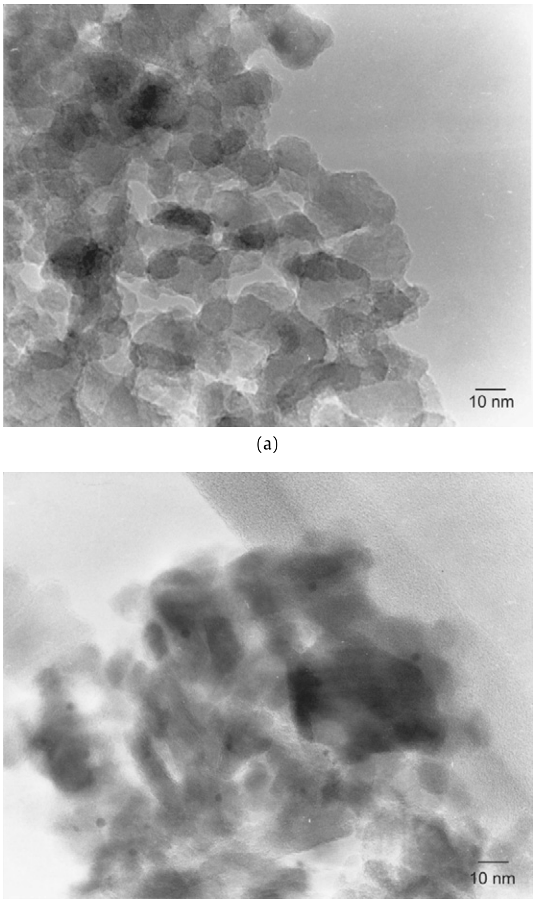


Fig. 3. Specific activities of different gold catalysts in glucose oxidation. Reaction conditions: 40 °C, pH 9, 100 $mmol\,l^{-1}$ glucose.

The catalyst prepared with 2 M HCl showed the highest activity. Similarly high activity was obtained for the catalyst prepared with nitric acid 2 M HNO_3 and 2 M NaCl. For those two catalysts, the hydrogen tetrachloroaurate complex likewise was the dominant species during catalyst preparation. In contrast, the catalyst prepared with 0.25 M NaOH, in which the tetrachloroaurate complex was completely diminished, exhibited the lowest activity. Thus, it can be assumed that stabilization of the tetrachloroaurate complex in the catalyst precursor is the key step in the catalyst preparation procedure to obtain a catalyst with high activity. The gold catalysts, in which the tetrachloroaurate ion was stabilized in the impregnation solution, were, with 1880 $mmol\,min^{-1}\,g_{Au}^{-1}$, twice as active in glucose oxidation than the gold catalyst prepared with water, and thus also were more active than the DP catalysts. Thus, these catalysts were more active than the gold catalysts used so far for glucose oxidation and thus can provide even wider industrial applicability.

TEM analysis of the most active catalyst ($HAuCl_{4(s)}$ 2 M HCl) and a less active catalyst ($HAuCl_{4(aq)}$ H_2O) was carried out (Fig. 4).



(b)

Fig. 4. TEM pictures of the gold catalyst prepared from the impregnations solutions: (a) $HAuCl_{4(s)}$ 2 M HCl and (b) $HAuCl_{4(s)}$ H_2O.

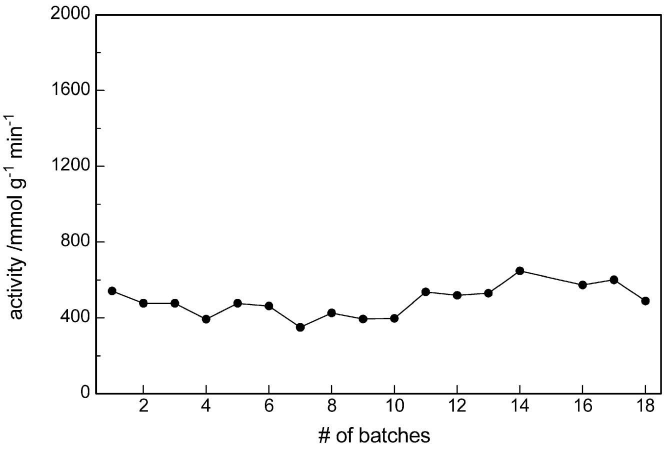


Fig. 5. Activity of the repeated batches during the long-term stability test of the catalyst $HAuCl_{4(s)}$ 2 M HCl. Reaction conditions: 40 °C, pH 9, 30 wt% glucose.

The $HAuCl_{4(s)}$ 2 M HCl catalyst exhibited very small gold particles 1–2 nm in diameter, whereas the $HAuCl_{4(aq)}$ H_2O catalyst had gold particles 2–5 nm in diameter. Although both catalysts had only gold particles of sizes known to be active in glucose oxidation [25], the $HAuCl_{4(s)}$ 2 M HCl catalyst with the smaller particles is considerably more active. It is reasonable to assume that the greater activity can be attributed to the higher dispersion of the small gold particles, which result directly from the chloride-stabilized precursors.

The result that gold precursors containing chloride were suitable for the preparation of active gold catalysts were apparently in contrast with the previous literature. The presence of chloride during the preparation of gold catalysts is known to promote the agglomeration of small gold particles leading to large, and thus inactive, ones [15,26]. But in these previous studies, the catalysts were activated by calcination. In the present work, the activation of the gold catalysts was carried out by gas-phase reduction with hydrogen, and the catalysts showed excellent activity and selectivity toward gluconic acid in glucose oxidation. The results indicate that reduction with hydrogen is a good method for overcoming the disadvantages due to the presence of chloride during catalyst preparation. For the liquid-phase glucose oxidation considered here, residual chloride, if still present after catalyst preparation, would not affect the catalyst's properties, because it would be immediately dissolved in the reaction suspension.

For an efficient industrial process, the catalyst must exhibit not only high selectivity and activity, but also good long-term stability under industrially relevant conditions. Therefore, the most active catalyst, $HAuCl_{4(s)}$ 2 M HCl (0.3 wt% Au/Al_2O_3), was tested in repeated batches at high glucose concentrations of 30 wt%. The catalytic activity is shown in Fig. 5. Because loss of catalyst occurs in each separation step, the starting amount of catalyst was high, accepting a slight mass transfer limitation in the first batches. Therefore, the average activity increased slightly through the batches. The selectivity toward sodium gluconate was >98% in each run. Fructose was the sole byproduct found. This was formed not by a gold-catalyzed process, but rather by the well-known base-catalyzed Lobry–de-Bruin–Alberda–van-Ekenstein rearrangement. ICP-AES analysis of the catalyst before the first batch and after the last batch showed no loss of gold during the experiment. After the 18th batch, the experiment was stopped, even though the catalyst still had good activity. During 18 repeated batches, no decline in activity was observed. Thus, the gold catalyst prepared with 2 M HCl showed very good long-term stability under closed industrial conditions.

4. Conclusion

We have described gold catalysts prepared by the incipient wetness impregnation method using hydrogen tetrachloroaurate dissolved in different aqueous solutions as an impregnation solution. Hydrogen tetrachloroaurate hydrolysis in aqueous solution and the composition of the gold complexes depended on the pH value and the chloride concentration. The catalyst precursors were activated by gas-phase reduction with hydrogen instead of the typically used calcination method. The gold catalyst prepared with a hydrochloric acid solution exhibited the greatest activity in glucose oxidation with 1880 $\text{mmol}\,\text{min}^{-1}\,g_{Au}^{-1}$. In fact, the investigations showed that high chloride concentrations can be used to stabilize the tetrachloroaurate ion in the precursor solution, which then interacts strongly with the alumina support, leading to highly dispersed active gold catalysts. The gold particles had a mean diameter of 1–2 nm, and the activity was twice as high as that obtained for catalysts prepared by DP [16,27]. The TPR analysis of the catalyst precursors revealed a correlation between activity and TPR temperature. Catalyst precursors with higher TPR temperatures, and thus precursors with higher chlorinated gold complexes, showed a tendency toward higher activity. The gold catalyst prepared with 2 M HCl was tested in repeated batches at a glucose concentration of 30 wt%. No loss of activity was observed during 18 repeated batches. Thus, the prepared incipient wetness gold catalyst demonstrated very good long-term stability. In addition, industrial-scale catalyst preparation can be easily accomplished, because the incipient wetness method is a very facile process.

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