



M Ű E G Y E T E M 1 7 8 2

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Study of Gold Containing Bimetallic Catalysts in Selective Aerobe Oxidation of Alcohols

Thesis

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1. Introduction

For the sustainable development, the research of the more economic and eco-friendly chemical technologies, agreeing with the principal of green chemistry, are becoming more important and catalysis has an unquestionable role in it. Nowadays, the increasing demand of renewable resources, beside the fossil raw materials, makes a new challenge, that also calls for new catalytic processes.

The selective oxidation plays an important role in the fine chemical industry, like conversion of alcohols to higher value-added products. Thanks to Haruta¹ and Hutchings² pioneering works, an extensive research began in 1987, what confirmed the favorable properties of gold catalyst comparing even to the most effective noble metal catalysts. As heterogeneous catalysts usually gold nanoparticles are applied, which are stabilized on a support. The supports may play a direct role in the catalysis, so they can affect the selectivity or the activity, but their exact function has not been discovered yet. In gold catalysis - like usually in catalysis - bimetallic catalysts are used to further improve the performance. As a result, many high effective systems have been published in the literature, but many options left (e.g. changing the second metal and the structure of the particles) to optimize the catalyst in a given process. Our research group has already a long experience in gold catalysis especially in CO and glucose oxidation.

During my PhD work, I prepared mono and bimetallic gold (Au–Ag, Au–Ru, Au–Ir) nanoparticles and adsorbed them on oxide supports, then they were activated by different pretreatments. Their structures were studied by various complementary methods. The prepared catalysts were tested in benzyl alcohol and glycerol oxidation, furthermore several catalysts were tested in CO oxidation for comparison. My PhD work aim was to understand better the mechanisms of gold catalysis by searching correlations between the structure and the catalytic properties. I paid special attention to the effect of second metal and support as well as to the influencing factors which can modify these effects in the reaction.

¹ Masatake Haruta and others, *Chemistry Letters*, 16.2 (1987), 405–8.

² G. J. Hutchings, *Journal of Catalysis*, 96.1 (1985), 292–95.

2. Literature background

In the literature it is generally accepted that only the nanosize gold particles have activity in oxidation reactions, however, the ideal particle size is still disputed. The effect of particle size depends on the reaction, but often dissimilar ones were reported even in the same reaction. In CO oxidation the activity related to one surface gold atom typically increases with the decreasing size which is attributed to the increased concentration of low coordinated, more active Au atoms³. In selective oxidation similar phenomenon was also observed⁴, but some cases where the activity showed a maximum at certain size range⁵ were also published.

Supports are classified into two main categories. The “active” supports are typically reducible semiconductor oxides (e.g. TiO₂, Fe₂O₃), which help the activation of oxygen molecule by surface redox reaction⁶. The other group is the “passive” supports, where the non-reducible supports belong (e.g. Al₂O₃, SiO₂).

The oxidation of CO is a top priority reaction in terms of environment protection, medical and industrial usage. Several mechanisms have been proposed for gold catalysts, presumably several ones may prevail at the same time. In CO oxidation more mechanisms have been proposed. It can be assumed that more than one mechanism is present at one time. Some researchers suppose that the support is totally inactive in the reaction⁷, while others assume that the support or the vacancies on the support has a key role in the oxygen activation⁸. Regarding the active gold sites; some theories consider that only the metallic gold has activity⁹ while others suppose that the ionic gold (Au⁺ or Au³⁺) also takes part in the catalysis¹⁰. The activating effect of water content was examined in CO oxidation and the experiments showed that the water content determines the temperature profile of the catalysis¹¹.

Benzyl alcohol (BnOH) is a frequently used, easily oxidizable model molecule of selective oxidation catalytic tests. Moreover the obtained products (benzyl aldehyde, benzoic acid and

³ Ton V. W. Janssens and others, *Topics in Catalysis*, 44.1 (2007), 15.

⁴ Hironori Tsunoyama, Hidehiro Sakurai, and Tatsuya Tsukuda, *Chemical Physics Letters*, 429.4 (2006), 528–32.

⁵ Peter Haider and others, *Catalysis Letters*, 125.3–4 (2008), 169–76.

⁶ Masatake Haruta, *CATTECH*, 6.3 (2002), 102–15; C. K. Costello and others, *Applied Catalysis A: General*, 243.1 (2003), 15–24; Xiao Yan Liu and others, *Nano Today*, 8.4 (2013), 403–16.

⁷ Costello and others; Mitsutaka Okumura and others, *Applied Catalysis A: General*, Catalysis by Gold, 291.1 (2005), 37–44.

⁸ Haruta; Geoffrey C. Bond and David T. Thompson, *Gold Bulletin*, 33.2 (2000), 41–50.

⁹ Haruta; Mitsutaka Okumura and others, *Applied Catalysis A: General*, Catalysis by Gold, 291.1 (2005), 37–44.

¹⁰ Costello and others; Geoffrey C. Bond and David T. Thompson, *Gold Bulletin*, 33.2 (2000), 41–50

¹¹ D.A.H. Cunningham, W. Vogel, and M. Haruta, *Catalysis Letters*, 63.1 (1999), 43–47.

benzyl benzoate) have industrial importance. The byproduct, benzoic acid has a strong inhibition effect to the gold catalysts¹². This effect can be eliminated by adding base (as e.g. KF or K₂CO₃), and it can increase the activity. Promoting effect of water was demonstrated in a few studies¹³. For BnOH oxidation a mechanism was assumed, in which two gold atoms take part in the reaction at once and the surface OH assists in the deprotonation¹⁴.

The application of basic support was raised in BnOH oxidation¹⁵. Hydrotalcite was the most effective support which has both strong acidic and basic OH groups. Gold on supports having neither basic nor acidic centers showed a negligible activity.

After turn of the millennium the production of biofuels increased dramatically¹⁶. The process results in enormous amount of glycerol as byproduct, and its economical processing has not been solved up to now. One of the options is the selective oxidation of glycerol, but the number of the possible products is quite high (e.g. dihydroxyacetone, glyceraldehyde, tartronic acid). Thus, in this reaction the selectivity has also a great significance beside the activity. In general, the catalytic glycerol oxidation occurs at low temperature and in strongly basic, aqueous media. It was showed that with decreasing the size of the gold particles not only the activity changes, but the selectivity as well¹⁷.

Enhancing the activity, modifying the selectivity and increasing the resistance against inhibition can be usually achieved by applying a second metal. Gold and silver can form totally miscible solid solution. Thanks to this and the silver's relatively cheap price, it is one of the most studied bimetallic gold alloy catalysts. The structure of the particles depends on the preparation method. Activity enhancement was observed on almost every Au:Ag composition compared to monometallic ones in CO oxidation, moreover the silver improved the long-term stability¹⁸. On gold-silver catalysts at low silver content activity increase was observed in glucose¹⁹ and benzyl

¹² Emmanuel Skupien and others, *Catalysts*, 4.2 (2014), 89–115.

¹³ Emmanuel Skupien and others, *Catalysts*, 4.2 (2014), 89–115.; Xiaomin Yang and others, *Catalysis Communications*, 9.13 (2008), 2278–81.

¹⁴ Emmanuel Skupien and others, *Catalysts*, 4.2 (2014), 89–115.

¹⁵ Wenhao Fang and others, *Chemistry – A European Journal*, 17.4 (2011), 1247–56.

¹⁶ A. Muscat and others, *Global Food Security*, 2019, 100330.

¹⁷ William C. Ketchie and others, *Journal of Catalysis*, 250.1 (2007), 94–101; S. Demirel-Gülen, M. Lucas, and P. Claus, *Catalysis Today*, 1st International Symposium on Carbon for Catalysis 'CARBOCAT-1', 102–103 (2005), 166–72; Elżbieta Skrzyńska and others, *Journal of Molecular Catalysis A: Chemical*, 382 (2014), 71–78.

¹⁸ Alberto Sandoval and others, *Journal of Catalysis*, 281.1 (2011), 40–49; Zhenping Qu and others, *Applied Surface Science*, 277 (2013), 293–301.

¹⁹ Tímea Benkó and others, *Applied Catalysis A: General*, 479 (2014), 103–11.

alcohol oxidation, in the latter the selectivity shifted towards to benzoic acid and benzyl benzoate in aqueous media²⁰.

According to a research of glycerol oxidation on Au–Ag/ZnO catalyst, which was published in 2018, silver decreased the activity compared to the monometallic gold catalyst²¹. It was explained on the one hand with the decreased gold surface, since silver spread on the gold surface, on the other hand silver increased the partial negative charge on gold which reduced the affinity of oxygen. Generally silver improved the activity of gold if the surface concentration of silver was not too high.

Au and Ru are immiscible in macroscopic scale. Though in nanoscale, different structures can be formed, depending on the support and the preparation method. On MgO supported sample Au cored particles were showed, with surface enrichment of Ru²². The synergic effect of Au and Ru was revealed in CO oxidation and total oxidation of methanol²³ as well as in selective oxidation of n-octanol and glycerol²⁴. This metal pair has not been tested in benzyl alcohol oxidation.

Among the three bimetallic systems investigated in my work the Au–Ir is the least studied. Iridium is not miscible at all with gold in bulk form. There is an article where the separation of two metals²⁵, but there is also an example where formation of alloyed nanoparticles were reported²⁶. In CO oxidation²⁷, in toluene complete oxidation and in reduction of cinnamic aldehyde²⁸ the bimetallic Au–Ir catalysts had advantageous properties as compared to the monometallic ones. This bimetallic system has not been examined in the selective oxidation of benzyl alcohol.

²⁰ Xuemin Huang and others, *Applied Catalysis A: General*, 467 (2013), 407–13.

²¹ Iveta Kaskow, Piotr Decyk, and Izabela Sobczak, *Applied Surface Science*, 444 (2018), 197–207.

²² S. Galvagno and others, *Journal of Catalysis*, 69.2 (1981), 283–91.

²³ Lina A. Calzada and others, *Applied Catalysis B: Environmental*, 207.Supplement C (2017), 79–92.

²⁴ Laura Prati and others, *Catalysis Science & Technology*, 1.9 (2011), 1624–29.

²⁵ Xim Bokhimi, Rodolfo Zanella, and Carlos Angeles-Chavez, *The Journal of Physical Chemistry C*, 114.33 (2010), 14101–9.

²⁶ Jia Zhao and others, *Catalysis Communications*, 54.Supplement C (2014), 72–76.

²⁷ Alejo Aguirre and others, *Topics in Catalysis*, 59.2–4 (2016), 347–56.

²⁸ Jia Zhao and others, *Catalysis Communications*, 54.Supplement C (2014), 72–76.

3. Experimental

Preparation of the catalysts

Catalyst labeling used: for example in Au₈₅Ag₂₅/SiO₂-SOL the first part refers to the metal components and their concentration (85 $\mu\text{mol Au} + 25 \mu\text{mol Ag} / \text{g}_{\text{cat}}$), the next part shows the applied support, while the part after hyphen displays the preparation method. According to the SOL method the metal particles were formed by reducing the metal ions of the precursor (in case of bimetallic particles by consecutive- or co-reduction of the precursors) with NaBH₄ in presence of polyvinyl alcohol (PVA) stabilizing agent. Then the prepared sol particles were adsorbed on the support. The Ir/Al₂O₃ sample was made by impregnation. Au–Ag bimetallic and the corresponding monometallic particles were fabricated also by SMAD method. In this method the vapors of the metal or two metals were co-condensed with acetone vapor on the cold (–197 °C) walls of a static glass reactor. After melting the solid matrix the resulting solvated metal atoms (SMA) solution was adsorbed on the support. Usually the catalyst samples were examined after calcination (at 400 °C/air) and also after a following reduction (at 350 or 400 °C/H₂) pretreatment.

Characterization techniques

The acid-base properties of the supports were characterized by temperature programmed desorption (TPD) of CO₂ and NH₃. The metal contents of the catalysts were determined by prompt- γ activation analysis (PGAA) or ICP–OES measurements. The size and the elemental composition of the particles were examined with transmission electron microscopy (TEM) equipped with EDS. X-ray photoelectron spectroscopy (XPS), temperature programmed reduction (TPR), CO adsorption followed by diffuse reflectance infrared Fourier transformation spectroscopy (CO–DRIFTS) and CO+Kr pulse chromatography were used to characterize the metal particles of the catalysts.

Catalytic investigations

The temperature programmed CO oxidation tests were performed with 0.5% CO + 10% O₂ in He reactant mixture at atmospheric pressure in a plug flow reactor connected to a mass spectrometer (QMS) for analysis.

The benzyl alcohol oxidation was performed in toluene or xylene solution (1 M or 0.1 M benzyl alcohol) at 80 or 60 °C with and without added K₂CO₃, the oxygen was bubbled through the

solution. The analysis was made with GC device equipped with a FID detector. The reaction rate was determined from the initial, nearly linear part of the conversion versus reaction time curves.

The glycerol oxidation tests were carried out in 0.3 M aqueous solution of glycerol at 50 °C in presence of 1.2 M NaOH, in a glass reactor under 3 bar O₂ pressure. The analysis of the reaction mixture taken at regular time intervals was executed with a HPLC and the components were detected with UV and refractive index (RI) detector.

4. Results

Comparing Au–Ag/SiO₂ catalysts in glucose, benzyl alcohol and CO oxidation

Different Au:Ag molar ratio silica supported catalysts had been created with SOL method and were tested in glucose oxidation after calcination and the after following reduction by my co-workers²⁹. In my work, I tested these catalysts in base-free selective benzyl alcohol oxidation and CO oxidation, and these data were compared with the earlier glucose oxidation results.

Both the Au:Ag ratio and the pretreatment affected the active temperature region and the reaction rate of CO oxidation. The gold-containing catalysts displayed high activity in two well separable, lower and higher temperature ranges (LT and HT, Fig. 1). This implies that the CO oxidation has at least two different reaction mechanisms which have different active temperature range. The reduction increased the HT activity, but usually decreased the LT activity. From this, I can conclude that the two mechanisms have different active sites and the reduction favored the HT active site formation. Moreover, the low temperature active site has to include gold atom because the Ag₉₂/SiO₂-SOL had a low activity in the LT range.

The activity order of the samples was alike in benzyl alcohol and glucose selective oxidation as well as in case of the calcined samples in CO oxidation. In the base-free BnOH oxidation, the catalysts were deactivated at low conversions. The high Au content bimetallic samples exhibited the highest initial activity, while the Au₄₉Ag₅₁/SiO₂-SOL with rather small Au surface and the monometallic silver were inactive in the selective oxidations both in calcined and reduced states.

Surprisingly, the calcined Au₁₉₀/SiO₂-SOL was also proved to be inactive in benzyl alcohol oxidation, but after reduction it had a medium activity. Small quantity of silver improved the activity of gold but the further silver addition led to deactivation. The inactivity of

²⁹ Tímea Benkó and others, *Applied Catalysis A: General*, 479 (2014), 103–11.

Au49Ag51/SiO₂-SOL is supposed to be due to the absence of sites with enough number of neighboring surface Au atoms to adsorb the substrate, just like it was explained in case of glucose oxidation³⁰. The activating effect of reduction was much stronger in benzyl alcohol oxidation than in glucose oxidation.

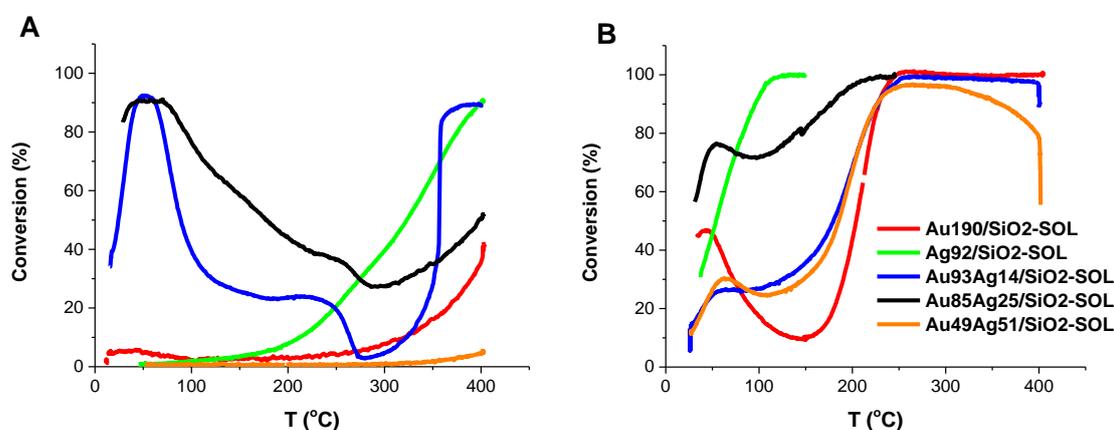


Fig. 1. Temperature programmed CO oxidation conversion curves of bimetallic AuAg/SiO₂ and the corresponding monometallic catalysts in the 2nd run on the calcined (A) and the 1st run on reduced samples (B)

Oxidation of glycerol over bimetallic Au–Ag/Al₂O₃ catalyst

Al₂O₃ supported Au–Ag catalysts (Au:Ag = 4:1) were prepared by two different methods (SOL and SMAD). The presence of bimetallic particles and surface enrichment of silver were revealed by STEM–EDS and XPS measurements. The SOL samples had better stability against sintering than SMAD ones, which indicates the protective effect of the stabilizer residues. The XPS measurements showed larger surface enrichment of silver on the bimetallic SMAD sample than on the SOL one, and the calcination increased it further. On the surface of the as prepared bimetallic samples, the silver was in metallic state, while on the monometallic silver it was partially oxidized. After calcination the oxidation state of silver did not change on the bimetallic SOL catalyst, however a little oxidation was observed on the SMAD one. This must be in relation with the weaker Ag–Au interaction on the surface having thicker Ag layer in the latter case.

The monometallic silver samples had a negligible activity in glycerol oxidation. The much higher activity of gold-containing SOL catalysts compared to the SMAD ones was caused by the higher dispersion and lower extent coverage of gold by silver in the SOL samples. Addition of silver increased the SOL and significantly enhanced the SMAD samples' activity. The increased

³⁰ Tímea Benkó and others, *Applied Catalysis A: General*, 479 (2014), 103–11.

activity of the calcined SOL catalysts related to the as prepared ones was the result of the removal of the stabilizer's residues. After calcination the synergetic effect decreased especially on the SMAD sample, where fully vanished, what could be trace back to the enhanced surface enrichment of silver. Silver addition promoted the further oxidation of glyceric acid to tartronic acid on the SOL sample. This phenomenon was not observed on the less active SMAD catalysts.

Benzyl alcohol oxidation over bimetallic Au–Ru/Al₂O₃ and Au–Ir/Al₂O₃ catalysts

On both bimetallic samples the surface enrichment of the second metal was confirmed by structural investigations on the bimetallic particles. The interaction with gold was also proved by the modifications of the redox properties of Ru and Ir. As the monometallic iridium and ruthenium has a negligible activity in BnOH oxidation compared to gold, thus I think that on the bimetallic catalysts the catalysis took place mostly on the gold surface, which was modified or complemented by the cooperating Ru or Ir as e.g. in oxygen activation.

On the bimetallic samples an induction period appeared in the catalytic tests with added K₂CO₃. This might be caused by the contaminations (rather water) adsorbed on Ru and Ir from air, which affects the gold surface as well, so it hindered the adsorption of the reactants. Following the induction period the reaction rate of the bimetallic Au–Ru sample was almost as high as the reaction rate of the monometallic Au one (Fig. 2). The reduction significantly increased the activity of the calcined iridium containing samples. The reduced bimetallic Au–Ir catalyst had a relatively high activity but it was lower than the activity of the bimetallic Au–Ru catalyst. Based on the estimated activity related to surface Au atoms, there is a synergetic effect between Au and Ru, and in reduced state also between Au and Ir. Metallic ruthenium and iridium chemisorb oxygen dissociatively, and by that may supply more reactive oxygen to regenerate the Au centers from Au–H forming during the β -hydride elimination. After calcination the presence of ruthenium and iridium increased and stabilized the partial positive charge of gold (Au ^{δ^+}), what may help the substrate adsorption and β -hydride elimination.

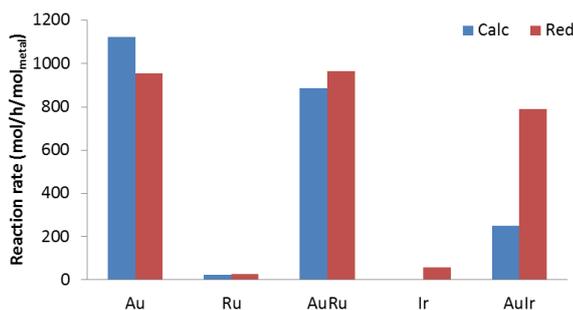


Fig. 2. The initial activity of Au91/Al₂O₃-SOL, Ru49/Al₂O₃-SOL, Au47Ru35/Al₂O₃-SOL, Ir187/Al₂O₃-IMP and Au49Ir15/Al₂O₃-SOL in benzyl alcohol oxidation (0.1 M BnOH and K₂CO₃ in toluene, 80 °C)

Support effect in benzyl alcohol oxidation

Based on the CO₂- and NH₃-TPD measurements, among of the five tested, non-reducible supports, SiO₂ had neither acidic nor basic centers, MgO had only basic centers, while Al₂O₃, MgAl₂O₃ and HAP contained both basic and acidic centers, but their amount and strength were different. The surface OH concentration was also disparate on the supports. The calcination at 400 °C resulted in different degrees of sintering of Au particles on the different supports. The CO adsorption followed by DRIFT spectroscopy showed that in calcined catalysts the gold had a partial positive charge which quantity and stability depended on the support. The co-adsorption of oxygen induced smaller, but also varying extent electron deficiency on the metallic Au. The amount of reversibly adsorbed CO was quantitatively determined by CO+Kr pulse chromatography followed by mass spectrometer, which correlated well with the relative intensity of IR bands of adsorbed CO.

The benzyl alcohol oxidation in xylene without added base stopped at low conversions due to the inhibition of benzoic acid. Adding K₂CO₃ ceased this effect and significantly increased the catalysts activity (Fig. 3). The selectivity of benzaldehyde was sufficient (>75%) even at high conversion, but the acidic centers slightly increased the production of benzyl benzoate. Based on the reaction rates related to surface gold atoms the strongest basic centers containing MgO and MgAl₂O₄ supported catalysts were the most actives. The HAP support containing high concentration, but weaker basic centers had less activity, than was expected based on the Au particle size, what was explained by the less accessibility of gold surface. The lack or low concentration of basic centers resulted in little activity on the SiO₂ and the Al₂O₃ supported catalysts, respectively. The Au/SiO₂ sample was inactive without K₂CO₃, which is similar to my previous result, so the Au nanoparticles alone are not enough to catalyze the BnOH oxidation in the applied circumstances.

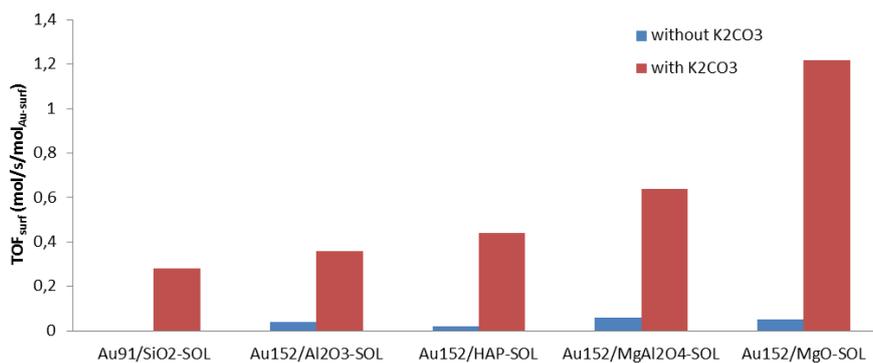


Fig. 3. Surface gold atoms related initial activity of different supported Au catalysts with and without added K₂CO₃ (0.1 M BnOH in xylene, 60 °C)

These results emphasize that the basicity of the support favors the activity. However, the Au dispersion, the accessibility of the gold surface, the acidic-basic character, the OH groups and H₂O concentration of the support, and the added base determine together the behavior of gold NPs in BnOH selective oxidation.

5. Thesis

1. On Au–Ag/SiO₂ catalysts the CO oxidation occurs via at least two different mechanisms, which have different active temperature range. The reaction changes mechanism with increasing temperature. The two mechanisms have different active centers. The hydrogen reduction prefers the formation of the active centers of high temperature mechanism. Furthermore, gold atoms are needed to form the low temperature centers. (1)
2. The presence of nanoscale gold particles alone is not a sufficient condition of the benzyl alcohol oxidation in organic solvent. The presence of a proper promoter is also needed (basic surface sites or added base, water, second metal) for the catalysis. Promoters can assist to deprotonation of benzyl alcohol (basic centers, water), modify the charge state of Au centers (second metal, acid-basic properties of support) helping the adsorption of substrate and the β -hydride elimination, contribute oxygen activation (second metal), which favors the regeneration of the Au centers. The quality of the OH groups affects the selectivity. (1, 3)
3. There is a strong synergetic effect between gold and silver on SiO₂ support in base-free benzyl alcohol oxidation, if on the surface of bimetallic particles the concentration of gold atoms is larger. It implies that the adsorption of the substrate has a necessary minimum contiguous gold surface, as it was suggested in case of glucose oxidation. The trend of Au:Ag atomic ratio-dependent change in catalytic activity is similar to that previously published in glucose oxidation, what indicates the similarity of the active centers in the two reactions. (1)
4. Adding silver to gold (Au:Ag = 4:1 atomic ration) improves the activity of the AuAg/Al₂O₃ catalysts made by sol adsorption (SOL) and solvated metal atom deposition (SMAD) method in the selective oxidation of glycerol. The lower activity of the SMAD catalyst related to the SOL one can be attributed to its larger particle size and the higher surface enrichment of silver on its bimetallic particles. On the SOL derived catalyst it was demonstrated first time that silver promotes the further oxidation of glyceric acid to tartronic acid. (2)
5. Au–Ru and Au–Ir bimetallic nanoparticles were made in aqueous media in a way, which have not been used before, Au and Ru, or Au and Ir precursor ions were co-reduced by NaBH₄ in presence of PVA stabilizer. According to the STEM–EDS examinations the modifier metal (Ru, Ir) enriches on the bimetallic particle's surface. The structural

investigations suggested that this structure remains on the aluminum-oxide support after calcination and following reduction at 400 °C. (4)

6. Based on the activity related to the unit of the total molar amount of the metals, the Au–Ru/Al₂O₃ and the reduced Au–Ir/Al₂O₃ were somewhat less active than the Au/Al₂O₃ in benzyl alcohol oxidation in presence of base (K₂CO₃), while the Ru/Al₂O₃ and the Ir/Al₂O₃ had a negligible activity. However, based on the comparison of the estimated activity related to unit of the surface Au atoms, a synergetic effect between Au and Ru, and in reduced state also between Au and Ir is suggested. (4)

6. Possible applications

The Au–Ag bimetallic catalysts on SiO₂ support have a good CO oxidation activity even at low temperature. This ability has great industrial potentials. One of the most important applications is the quality improvement of air. It is essential, where ventilation is not possible, only recycling of air, as on submarines or on spaceships. Another big research field is the automotive catalytic converters, where gold containing catalyst bed is used in the second step, so it contacts with a colder gas, where it has better performance, than the common platina/palladium catalysts. The bimetallic Al₂O₃ supported Au–Ag catalyst made by SOL method significantly improved the yield of tartronic acid in glycerol oxidation, which is more valuable than glycerol or glyceric acid. So this catalyst has a great opportunity in the exploitation of glycerol, the byproduct of the biofuel production. The selective alcohol oxidation activity showed by the catalysts investigated has high significance in the fine chemical industry (e.g. producing aldehydes), since it makes possible a much “greener” production compared to conventional manufacturing processes.

7. Publication

Related to the Theses

1. **Gergely Nagy**, Tímea Benkó, László Borkó, Tamás Csay, Anita Horváth, Krisztina Frey, Andrea Beck

Bimetallic Au–Ag/SiO₂ Catalysts: Comparison in Glucose, Benzyl Alcohol and CO Oxidation Reactions

Reaction Kinetics, Mechanisms and Catalysis, 115 (2015) 45–65

DOI: 10.1007/s11144-015-0835-2, IF: 1.265 FI: 15

2. Jouve Andrea, **Gergely Nagy**, Ferenc Somodi, Cristina Tiozzo, Alberto Villa, Antonella Balerna, Andrea Beck, Claudio Evangelisti, Laura Prati

Gold-Silver Catalysts: Effect of Catalyst Structure on the Selectivity of Glycerol Oxidation

Journal of Catalysis, 368 (2018) 324–35

DOI: 10.1016/j.jcat.2018.10.019, IF: 7.723 FI: 2

3. **Gergely Nagy**, Andrea Beck, György Sáfrán, Zoltán Schay, Shaofeng Liu, Tianbo Li, Botao Qiao, Junhu Wang, Károly Lázár

Nanodisperse Gold Catalysts in Oxidation of Benzyl Alcohol: Comparison of Various Supports under Different Conditions

Reaction Kinetics, Mechanisms and Catalysis, 128 (2019) 71-95.

DOI: 10.1007/s11144-019-01615-8, IF: 1.520 FI: 3

4. **Gergely Nagy**, Gál Teodóra, Ferenc Dávid Srankó, György Sáfrán, Boglárka Maróti, István Sajó, Franz Schmidt, Andrea Beck

Selective Aerobic Oxidation of Benzyl Alcohol on Alumina Supported Au-Ru and Au-Ir Catalysts

Molecular Catalysis, 492 (2020), 110917

DOI: 10.1016/j.mcat.2020.110917, IF: 3.678 (2019) FI: 0

Other publications

5. Marta Stucchi, Andrea Jouve, Alberto Villa, **Gergely Nagy**, Miklós László Németh, Claudio Evangelisti, Rodolfo Zanella, Laura Prati

Gold-Silver Catalysts: Ruling Factors for Establishing Synergism

ChemCatChem, 11 (2019), 4043-4053

DOI: 10.1002/cctc.201900591, IF: 4.853 FI: 0

Oral presentations

1. **Gergely Nagy**, Dávid Srankó, László Borkó, György Sáfrán, Geszti Tamásné, Andrea Beck
Kétfémes Au-Cu/Al₂O₃ katalizátorok: szinergikus hatás benzil-alkohol aerob szelektív oxidációjában

MKE 2, (2015), Hajdúszoboszló

2. **Gergely Nagy**; Andrea Beck; Tímea Benkó; György Sáfrán; Dávid Srankó
Aluminum oxide-supported bimetallic Au-Cu catalysts in the selective aerobic oxidation of glucose and benzyl alcohol

V. Interdiszciplináris Doktorandusz Konferencia, (2016), Pécs

3. **Gergely Nagy**

Kétfémes katalizátorok aerob szelektív oxidációban

Fiatal Kutatók Napja, (2016), Budapest

Posters

1. **Gergely Nagy**, György Sáfrán, Dávid Srankó, László Borkó, Zoltán Schay, Boglárka Maróti, Franz Schmidt, Ferdinand Hofer, Andrea Beck

Aerobic oxidation of benzyl alcohol and glucose on alumina supported Au-containing bimetallic catalysts: Comparison of Ag, Cu, Ru and Ir as second metals

13th Pannonian International Symposium on Catalysis (2016), Siófok

2. **Gergely Nagy**, Ferenc Somodi, György Sáfrán, Zoltán Schay, Teodóra Gál, Andrea Beck, Laura Prati, Claudio Evangelisti, Cristina Tiozzo

Sol derived alumina and silica supported Au-Ag bimetallic catalysts: structure and activity in aerobic selective oxidation of benzyl alcohol

33rd European Conference on Surface Science (ECOSS 33) (2017), Szeged

3. **Gergely Nagy**, Dávid Srankó, Károly Lázár, György Sáfrán, Andrea Beck, Shaofeng Liu, Tianbo Li, Hailian Tang, Botao Qiao, Rile Ge and Junhu Wang

Support effect of Au catalysts in aerobic benzyl-alcohol oxidation: effect of acid-base properties

14th Pannonian International Symposium on Catalysis (2018), Starý Smokovec, Slovakia