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Solvent-free selective oxidation of benzyl alcohol to benzaldehyde by *tert*-butyl hydroperoxide over U_3O_8-supported nano-gold catalysts

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ABSTRACT

Liquid-phase oxidation of benzyl alcohol to benzaldehyde by tertiary butyl hydroperoxide over different uranium oxide supported nano-gold catalysts in the absence of a solvent has been thoroughly investigated. The influences of catalyst parameters [viz. method of gold deposition (impregnation, co-precipitation, deposition–precipitation and homogeneous deposition–precipitation), gold loading (0–8 wt%) and catalyst calcination temperature (100–900 °C)] on the catalyst performance have been investigated. The influence of reaction conditions [viz. reaction time (0–2 h) and temperature (25–94 °C)] on the process performance has also been studied. The Au/U_3O_8 catalyst prepared by the homogeneous deposition–precipitation and calcined at 400 °C showed very high activity (100% benzyl alcohol conversion with >85% selectivity for benzaldehyde) in the process for a short reaction period (0.5 h) at 94 °C. The catalyst also showed excellent reusability in the process.

1. Introduction

Benzaldehyde is an important intermediate for the production of perfumes, pharmaceuticals, dyestuffs and agrochemicals. It is commercially produced as a by-product of the oxidation of toluene to benzoic acid [1]. It can also be produced by the hydrolysis of benzal chloride. However, the latter process provides benzaldehyde with trace contamination from chlorine which is not acceptable in perfumes and pharmaceuticals. With regards to chlorine-free benzaldehyde production, several studies have been reported on the catalytic vapor-phase oxidation of benzyl alcohol to benzaldehyde [2–7]. However, in the vapor-phase oxidation process, a very significant carbon loss in the form of carbon oxides is a major problem. A few studies have, therefore, also been reported on the liquid-phase oxidation of benzyl alcohol to benzaldehyde (which is considered as a model oxidation reaction) over the palladium [8,9], Ni–Al hydrotalcite [10] and heteropolyacid [11] catalysts in the presence of different solvents. Recently, the use of MnO_4^{-1}-exchanged Mg–Al hydrotalcite [12] and other transition metal containing hydrotalcite-like solids [13,14], as an active/selective catalyst for the solvent-free oxidation of benzyl alcohol to benzaldehyde, using TBHP and O_2 as an oxidizing agent have also been reported.

After the pioneering work of Prof. Haruta [15], gold has attracted the attention of a number of investigators for oxidation/reduction reactions. Galvagno and co-workers [16,17] and Rossi and co-workers [18,19] reported oxidation of alcohols to aldehydes using Au/Fe_2O_3 and Au/SiO_2 catalysts, respectively. Recently, Choudhary et al. [20] reported a very good activity for a number of nano-gold catalysts, prepared by supporting gold on different metal oxides (such as alkaline earth, rare earth, group IIIa and transition metal oxides), for the solvent-free oxidation of benzyl alcohol to benzaldehyde by molecular O_2. Among the supported nano-gold catalysts (prepared by homogeneous deposition method), the Au/U_3O_8 showed the best performance [20,21]. Later, Hutchings and co-workers [22,23] have reported the use of supported gold and Au–Pd catalysts for the solvent-free oxidation of primary alcohols to aldehydes by oxygen. It is preferable to use O_2 over hydrogen peroxide or organic hydroperoxide as an oxidizing agent for the benzyl alcohol-to-benzaldehyde oxidation. However, the benzaldehyde yield obtained is low, even under severe reaction conditions. Very recently, Wu et al. [24] have reported the use of hydrotalcite intercalated by sulphonated-salen–group (III) complexes for solvent-free oxidation of benzyl alcohol to benzaldehyde by H_2O_2. Although a number of studies have already been reported for the solvent-free oxidation of benzyl alcohol to benzaldehyde, either the conversion of benzyl alcohol or the selectivity for benzaldehyde has been low. Hence, there is a need to develop a more active and selective catalyst and/or a more environmentally benign process for the oxidation of benzyl alcohol.

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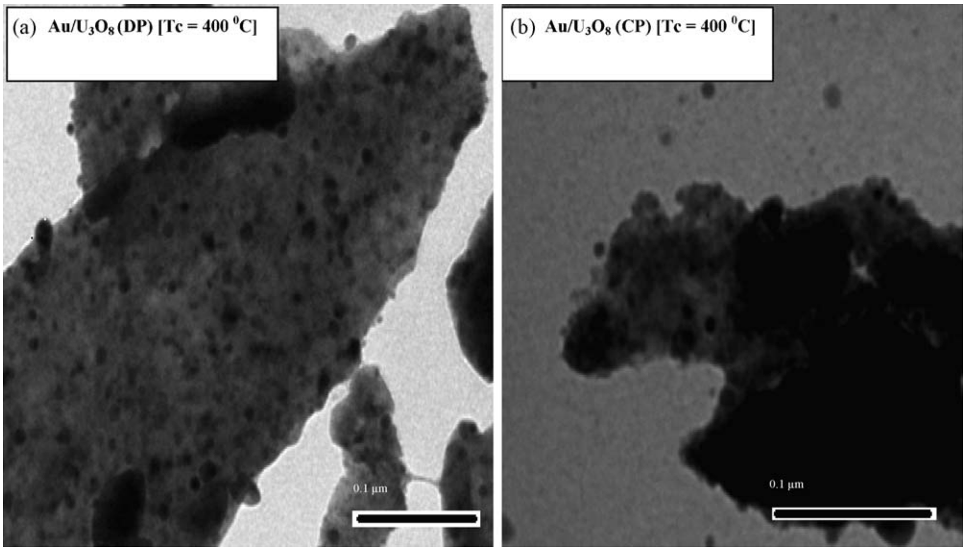

Fig. 1. TEM photographs of (a) Au/U_3O_8 (DP) and (b) Au/U_3O_8 (CP) (both calcined at 400 °C).

Our earlier work [20,21] revealed that among the different metal oxide supported gold catalysts the U_3O_8-supported catalyst showed the best performance in the benzyl alcohol-to-benzaldehyde oxidation (using O_2 [20] or TBHP [25] as the oxidizing agent). Earlier, U_3O_8 was used as a catalyst for the oxidative destruction of chloro organic compounds [26]. In this paper we report that benzyl alcohol can be completely converted with high selectivity for benzaldehyde in the solvent-free oxidation of benzyl alcohol by aqueous TBHP over Au/U_3O_8 catalyst [prepared by the homogeneous deposition–precipitation (HDP) method] at shorter reaction periods (∼0.5 h) and also under milder conditions (at <100 °C) than those employed earlier. The influences of different catalyst parameters (viz. method of gold deposition, Au loading and catalyst calcination temperature) and reaction conditions (viz.

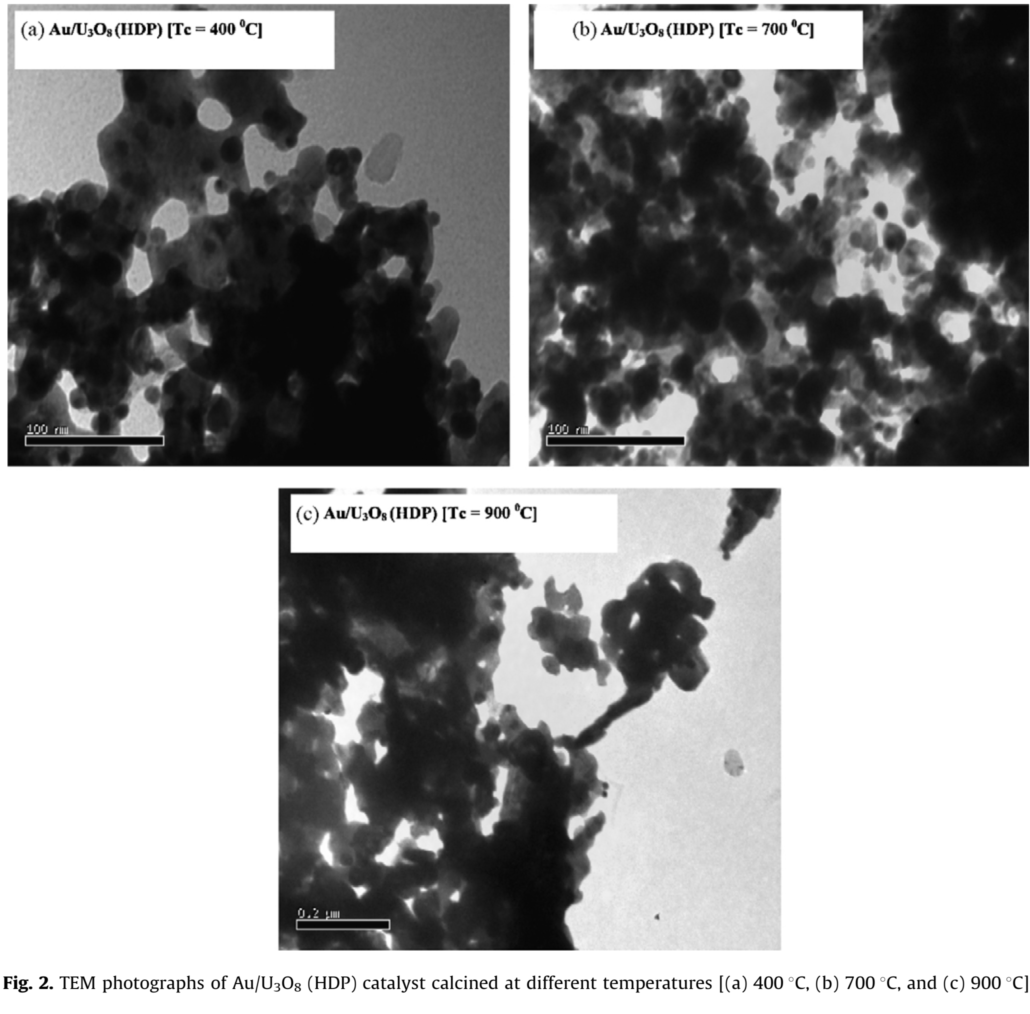

Fig. 2. TEM photographs of Au/U_3O_8 (HDP) catalyst calcined at different temperatures [(a) 400 °C, (b) 700 °C, and (c) 900 °C].

catalyst concentration and reaction period and temperature) on the process performance have also been exhaustively investigated.

2. Experimental

Preparation of Au/U$_3$O$_8$ catalysts by different gold deposition methods, such as the impregnation (IMP), deposition–precipitation (DP), homogeneous deposition–precipitation (HDP) and co-precipitation (CP) has been described earlier [21].

Unless otherwise mentioned, the catalysts were calcined at 400 °C for 2 h. The catalysts were characterized for their Au content (by ICP-OES), Au particle size [by the XRD peak (at $2\theta = 38.2°$) broadening and also by TEM] and surface area (by the single point N$_2$ adsorption method).

The liquid-phase oxidation of benzyl alcohol over the catalysts was carried out in a magnetically stirred round bottom flask (capacity: 25 cm^3), provided with a mercury thermometer (for measuring the reaction temperature) and a reflux condenser. Unless mentioned otherwise, the oxidation was carried out under the following reaction conditions: reaction mixture = 52 mmol benzyl alcohol + 78 mmol TBHP (70% TBHP in water) + 0.5 g catalyst, temperature = 94 °C and reaction time = 2 h. After the reaction, the catalyst was removed from the reaction mixture by filtration and the reaction products and benzyl alcohol were analyzed by gas chromatography with a flame ionization detector, using a SE-30 column and N$_2$ as a carrier gas. For measuring the catalyst performance for the oxidation as function of time, the liquid samples were removed intermittently from the reaction mixture by syringe and analyzed by the GC.

3. Results and discussion

The Au/U$_3$O$_8$ catalysts (prepared by different methods and calcined at different temperatures) were characterized for their Au loading, surface area and Au particle size. TEM photographs of the catalysts are shown in Figs. 1 and 2. XRD patterns of the catalyst support (U$_3$O$_8$) and Au/U$_3$U$_8$, prepared by the HDP method and calcined at different temperatures (100–900 °C) are presented in Fig. 3. The XRD patterns of the catalyst calcined at 100 °C and 300 °C are almost identical. Results of the catalyst characterization are presented in Table 1. The surface area of the Au/U$_3$O$_8$ (HDP) catalyst was decreased from 12 m^2 g^{-1} to 6 m^2 g^{-1} with increasing the Au loading from 1.1 wt% to 8.0 wt%.

Results showing the influence of the catalyst preparation method (viz. gold deposition on the support by DP, HDP, CP or impregnation) on the performance of the catalyst in the oxidation of benzyl alcohol are presented in Fig. 4. Figs. 5 and 6 show an influence of the Au loading and calcination temperature of the Au/U$_3$O$_8$ (HDP) catalyst on its performance in the benzyl alcohol oxidation. Influences of the process parameters (viz. reaction time and temperature) on the benzyl alcohol oxidation over the Au/U$_3$O$_8$ (HDP) catalyst are shown in Figs. 7 and 8.

Table 1
Properties of Au/U$_3$O$_8$ catalysts prepared by different methods.

Method of preparation	Calcination temperature (°C)	Au loading[a] (wt%) by ICP	Surface area (m^2 g^{-1})	Particle size of Au (nm)	
				TEM	XRD[b]
HDP	400	8.0	7	9.4	11.5
DP	400	5.1	8	15.5	17.2
Co-precipitation	400	4.7	5	16	17.5
Impregnation	400	8.0	6	34.1	38.4
HDP	700	8.0	6	20.3	22.4
HDP	900	8.0	5	32.3	34.3

[a] Amount of gold available for its deposition on support = 8.0 wt% of the support.
[b] From peak broadening of Au (2 0 0) peak ($2\theta = 38.5°$).

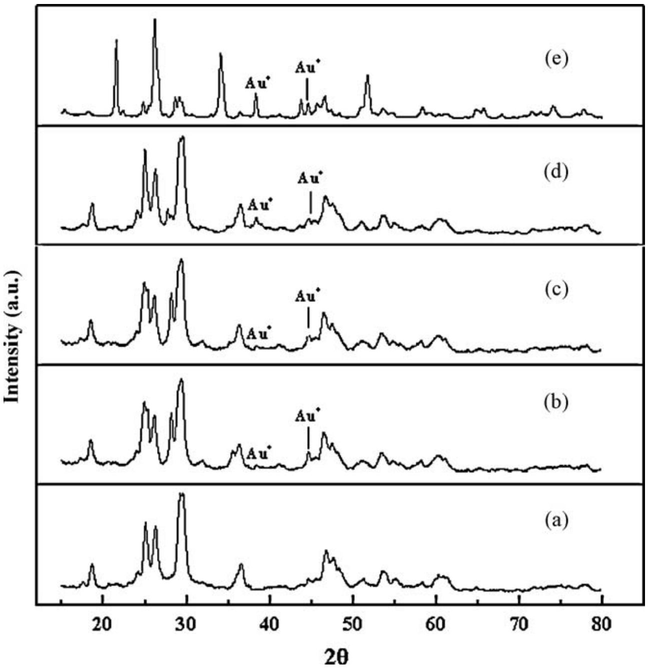

Fig. 3. XRD of U$_3$O$_8$ support calcined at 400 °C (a) and Au (8.0 wt%)/U$_3$O$_8$ (HDP) calcined at 100 °C (b), 300 °C (c), 400 °C (d) and 900 °C (e).

3.1. Influence of the method of gold deposition

Both the surface and catalytic properties of Au/U$_3$O$_8$ are strongly influenced by the method of gold deposition on the support (Table 1 and Fig. 4). The gold loading in case of the catalyst prepared by the incipient impregnation method is expected to be quantitative (i.e. 8.0 wt%). The HDP method also provides quantitative gold deposition (8.0 wt% Au) on the support. However, the Au loading of the catalyst prepared by the DP or CP method is much smaller (about 60% of the available gold). The Au particle size of the Au/U$_3$O$_8$ (HDP) is smallest while that of the Au/U$_3$O$_8$ (Imp.) is largest (Table 1). The gold particles in the catalysts prepared by the DP and CP methods are, however, intermediate in size.

Among the catalysts prepared by the different methods, the Au/U$_3$O$_8$ (HDP) showed the best performance (i.e. highest benzyl alcohol conversion and benzaldehyde yield) in the benzyl alcohol

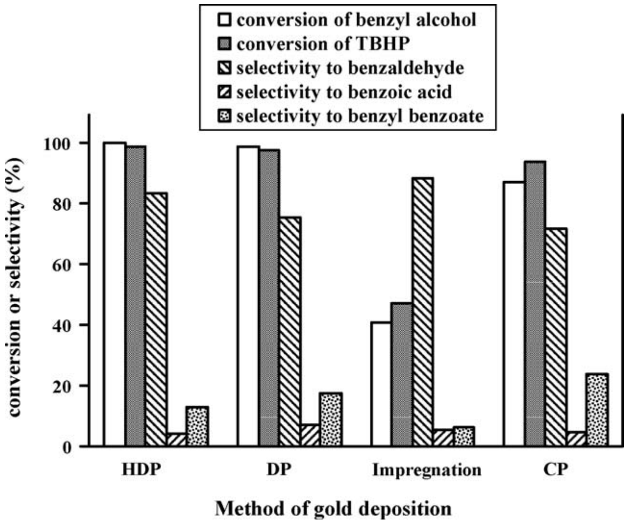

Fig. 4. Performance of Au/U$_3$O$_8$ catalysts prepared by different gold deposition methods and calcined at 400 °C in the oxidation of benzyl alcohol (amount of catalyst = 0.5 g, reaction temperature = 94 °C and reaction time = 2 h).

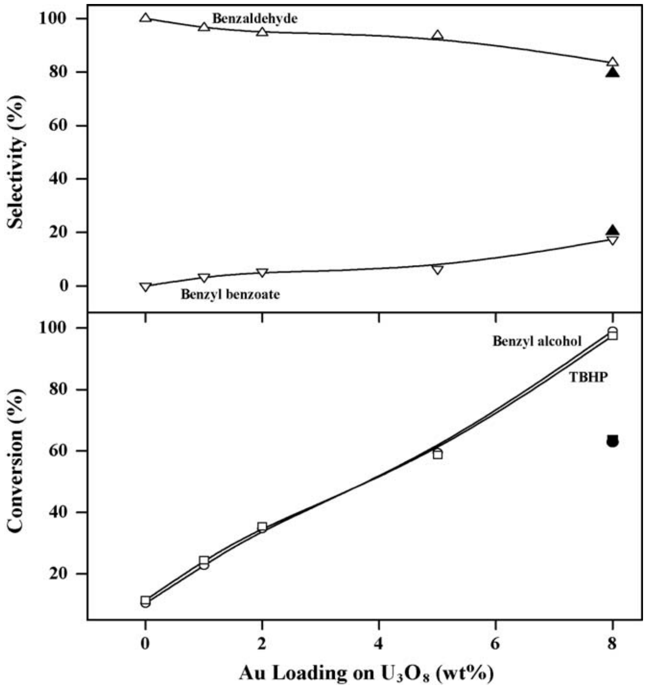

Fig. 5. Influence of the Au loading of Au/U_3O_8 (HDP) catalyst on its performance in the benzyl alcohol oxidation (amount of catalyst = 0.5 g, reaction temperature = 94 °C and reaction time = 2 h).

oxidation; the order of catalysts for their performance is as follows: Au/U_3O_8 (HDP) > Au/U_3O_8 (DP) > Au/U_3O_8 (CP) > Au/U_3O_8 (Imp.) (Fig. 2). This is consistent with the earlier studies which showed that the MgO, Yb_2O_3 or TiO_2 supported nano-gold catalyst (prepared by the HDP method) showed superior performance in the epoxidation of styrene [27–29].

The Au/U_3O_8 (HDP) and Au/U_3O_8 (Imp.) have similar Au loading (8.0 wt% Au) but the latter showed significantly poorer catalytic performance because of its larger Au particle size (Table 1). The Au/U_3O_8 (DP or CP) showed intermediate catalytic performance mostly because of its intermediate Au particle size and lower Au loading.

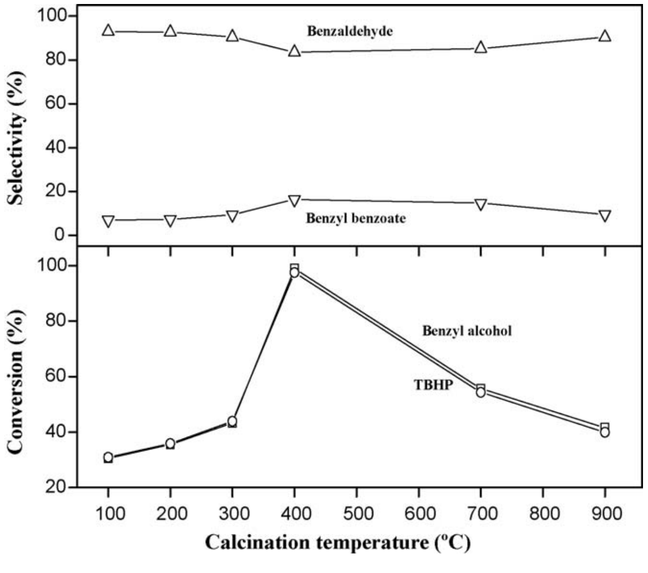

Fig. 6. Influence of calcination temperature of Au (8 wt%)/U_3O_8 (HDP) on its performance in the oxidation of benzyl alcohol (amount of catalyst = 0.5 g, reaction temperature = 94 °C and reaction time = 2 h).

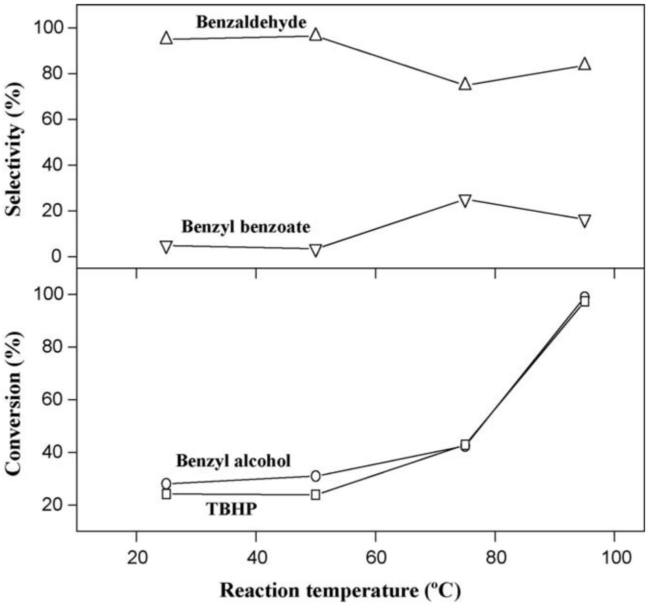

Fig. 7. Influence of reaction temperature on the performance of benzyl alcohol by TBHP to benzaldehyde over Au (8 wt%)/U_3O_8 (HDP) 400 °C (reaction time = 0.5 h).

The Au/U_3O_8 (HDP) showed excellent reusability in the benzyl alcohol oxidation; in the 1st, 3rd and 5th reuse of the catalyst, the benzyl alcohol conversion was 100% with benzaldehyde selectivity of 84%, 82% and 83%, respectively.

Since the Au/U_3O_8 catalyst prepared by the HDP method showed the best catalytic performance and excellent reusability, further detailed investigations on this catalyst were undertaken (described below).

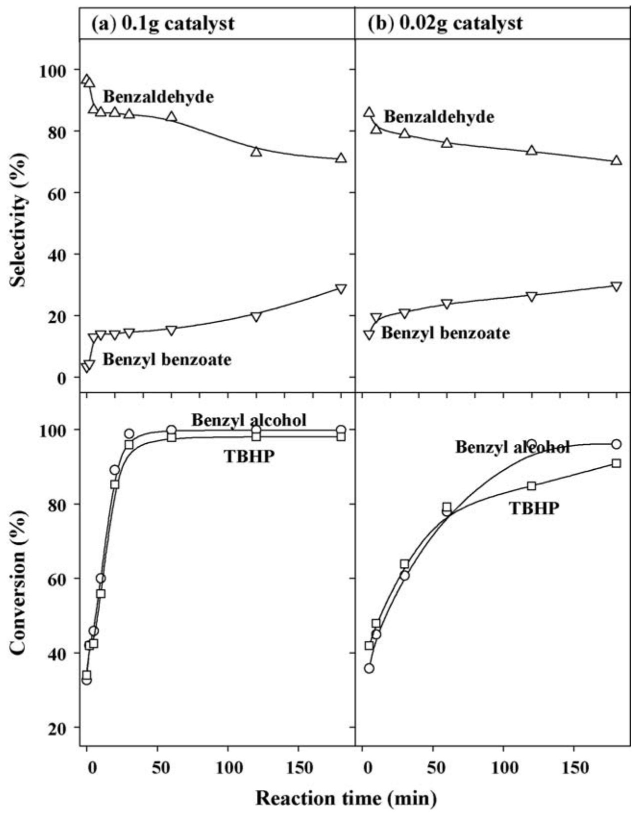

Fig. 8. Influence of reaction time and catalyst concentration on the performance of Au (8 wt%)/U_3O_8 (HDP) catalyst in the oxidation of benzyl alcohol (reaction temperature = 94 °C).

Table 2
Comparison of different heterogeneous catalysts for their performance in solvent-free liquid-phase benzyl alcohol-to-benzaldehyde oxidation by different oxidizing agents.

Catalyst	Oxidizing agent	Reaction conditions				Alcohol conversion (%)	BZH yield (%)	Productivity of BzH (mmol/g(cat)/h)	Reference
		Temperature (°C)	Pressure (atm)	Time (h)	Amount of catalyst[b] (g/mol)				
Ni–Al–HT[a]	O_2	90	1.0	12.0	250	–	31.0	0.1	[10]
Cu–Cr–HT	O_2	210	1.0	5.0	5.2	51.0	35.7	13.8	[13]
Au/U_3O_8	O_2	130	1.5	5.0	3.4	53.0	50.4	29.0	[20,21]
$Au–Pd/TiO_2$	O_2	100	10	0.5	0.3	1.9	1.6	105.0	[23]
$Au–Pd/Al_2O_3$	O_2	100	2	8.0	–	83.3	72.1	174.0	[22]
Au/TiO_2	O_2	100	2	8.0	–	15.3	9.8	240.0	[22]
S-S-Cr(III)–HT[c]	H_2O_2	50	1.0	2.0	6.0	65.8	65.8	54.8	[24]
$MnO_4{}^{-1}$-HT	TBHP	94	1.0	5.0	9.6	50.2	50.0	10.4	[12]
Co–Cr–LDH	TBHP	70	1.0	1.0	9.6	55.0	50.6	52.7	[14]
Au/MgO	TBHP	94	1.0	0.5	9.6	100	72.5	150.8	[30]
Au/TiO_2	TBHP	94	1.0	2.0	9.6	63.1	50.0	26.0	[31]
Au/U_3O_8	TBHP	94	1.0	0.5	1.9	100	85.0	884	Present work

[a] In the presence of toluene as a solvent.
[b] Weight in grams per mole of benzyl alcohol.
[c] Sulphonato-salen–Cr(III) hydrotalcite, HT = hydrotalcite.

3.2. Influence of Au loading in Au/U_3O_8 (HDP)

The results in Fig. 5 show a strong influence of the Au loading on the performance of benzyl alcohol oxidation.

As expected the conversion of benzyl alcohol increased with increasing Au loading; the increase was almost linear. However, the benzaldehyde selectivity decreased from 100% to 83% with increasing Au loading (0.0–8.0 wt%). The selectivity for benzyl benzoate showed the exactly opposite trend. The formation of benzoic acid was negligibly small. The support (U_3O_8) showed only a little benzyl alcohol oxidation activity (<10% conversion of benzyl alcohol, mainly to benzaldehyde).

On increasing the Au loading from 1.1 wt% to 8.0 wt%, the surface area of the catalyst decreased continuously from 12 $m^2\,g^{-1}$ to 6 $m^2\,g^{-1}$. The decrease in the surface area is expected mostly because of the adsorption of increasing amounts of chloride species on the catalyst support during the Au deposition for the higher gold loading in the catalyst.

3.3. Influence of catalyst calcination temperature

The performance of the Au (8.0 wt%)/U_3O_8 (HDP) in the benzyl alcohol oxidation is strongly influenced by its calcination temperature (Fig. 6). The benzyl alcohol conversion and benzaldehyde selectivity passed through a maximum and a minimum, respectively, with increasing calcination temperature. The change in the conversion is very large but that in the selectivity is relatively small. The benzaldehyde yield also passed through a maximum with increasing calcination temperature, indicating an optimum calcination temperature (400 °C) for the catalyst performance. The poor performance of the catalyst when it was calcined below 400 °C may be due to the presence of moisture in the catalyst. The sharp decrease in the oxidation activity of the catalyst, when calcined at higher than optimum temperature (i.e. above 400 °C), is mostly because of the growth (or sintering) of the gold particles. It may be noted that the Au particle size is increased from 9.4 nm to 32.3 nm with increasing the catalyst calcination temperature from 400 °C to 900 °C (Table 1). Our earlier studies [21] showed that the calcination temperature of the catalyst for its optimum performance in the oxidation of benzyl alcohol to benzaldehyde by molecular oxygen was also 400 °C.

3.4. Influence of reaction conditions

Results showing the influence of the reaction temperature, the time and also that of the amount of catalyst used on the process performance are presented in Figs. 7 and 8. Both the benzyl alcohol conversion and the product selectivities are strongly influenced by the reaction temperature, depending upon the reaction time (Fig. 7). As expected, the conversion increased exponentially when the reaction temperature was increased. However, the selectivity for benzaldehyde decreased and consequently that of benzyl benzoate also increased with increasing temperature. The influence of the reaction temperature is found to be very large above the temperature of 60 °C.

As expected the benzyl alcohol conversion increased with increasing reaction time and the time required for completing the benzyl alcohol oxidation was longer when a smaller amount of catalyst was used (Fig. 8). The selectivity for benzaldehyde is, however, decreased continuously with increasing reaction time. The exactly opposite trend is observed for the benzyl benzoate selectivity. The observed selectivity trends reveal the occurrence of following consecutive reactions in the benzyl alcohol oxidation:

$$C_6H_5{-}CH_2OH + (CH_3)_3COOH \rightarrow C_6H_5{-}CHO + (CH_3)_3COH + H_2O \quad (1)$$

$$C_6H_5{-}CHO + (CH_3)_3COOH \rightarrow C_6H_5{-}COOH + (CH_3)_3COH + H_2O \quad (2)$$

$$C_6H_5{-}COOH + C_6H_5{-}CH_2OH \rightarrow C_6H_5{-}COOCH_2C_6H_5 + H_2O \quad (3)$$

The formation of benzoic acid in the process was negligibly small and hence reaction (3) over the catalyst is expected to be quite fast.

3.5. Comparison of Au/U_3O_8 (HDP) with earlier catalysts

The performance (benzaldehyde productivity or turn over rate in solvent-free benzyl alcohol oxidation) of the Au (8.0 wt%/U_3O_8 (HDP)) catalyst is compared with that of the earlier reported active solid catalysts for the process in Table 2.

It may be noted that the Au/U_3O_8 (HDP) catalyst while using TBHP as the oxidizing agent (at 94 °C) showed highest productivity [0.9 mol/g(cat)/h], even at 100% conversion of benzyl alcohol, and very high yield (85%) for benzaldehyde. It is also noteworthy that the same catalyst while using molecular oxygen as an oxidizing agent showed much lower benzaldehyde productivity [0.03 mol/g(cat)/h] and yield (50%), even at a higher temperature (130 °C) [21].

The Au/MgO and Au/U_3O_8 catalysts have almost the same Au loadings (7.5 wt% and 8.0 wt%, respectively) and Au particle sizes

(9.0 nm and 9.4 nm, respectively). However, the Au/U_3O_8 catalyst showed much better performance in the benzyl alcohol-to-benzaldehyde oxidation (Table 2). This indicates the important role played by catalyst support in controlling the catalytic activity of supported gold catalyst through nano-gold particle–support-interactions.

For many years, uranium-based catalysts have been used in industrial practice and the procedures for their safe handling are well established [32]. However, since uranium is radioactive, long time exposure and/or inhalation of dust of uranium oxide should be avoided.

4. Conclusions

Nano-gold deposited on U_3O_8 by the homogeneous deposition precipitation method shows very high catalytic activity, selectivity and productivity and also shows excellent reusability in the solvent-free oxidation of benzyl alcohol to benzaldehyde by TBHP under mild reaction conditions. TBHP is a preferred oxidizing agent while using this catalyst in the benzyl alcohol-to-benzaldehyde oxidation for achieving 100% benzyl alcohol conversion (with very good benzaldehyde selectivity) in a short reaction period. The catalytic activity and selectivity are strongly influenced by the gold loading and the calcination temperature of the catalyst. The catalyst prepared by the DP, CP or impregnation method, however, showed inferior performance in the process.

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