Catal Lett (2009) 130:161–168
DOI 10.1007/s10562-009-9843-z

Alkylsilylated Gold Loaded Magnesium Oxide Aerogel Catalyst in the Oxidation of Styrene

Hadi Nur · Izan Izwan Misnon · Halimaton Hamdan

Received: 24 October 2008 / Accepted: 5 January 2009 / Published online: 30 January 2009

Abstract Aerogel-prepared magnesium oxide (A-MgO) was modified by attachment of gold particles. Subsequent modification of its surface with alkylsilylation of *n*-octadecyltrichlorosilane and chlorotrimethylsilane exhibits a high catalytic activity for oxidation of styrene with *tert*-butyl hydroperoxide in liquid phase. Catalytic results show that the introduction of gold on the surface of A-MgO gives a higher catalytic activity in styrene oxidation compared to titanium dioxide. The effect of surface modification of A-MgO was studied with XRD, UV–Vis DR, FTIR and nitrogen adsorption analysis.

Keywords Styrene oxidation · A-MgO · Gold · Titanium dioxide · Octadecyltrichlorosilane · Chlorotrimethylsilane

1 Introduction

It has been generally accepted that the overall chemical transformation in a catalytic reaction depend on the nature of the active site; its accessability and surroundings. Therefore, the important properties of a heterogeneous catalyst are an active site with the correct ensemble of metal atoms, metal ions, or other active components such as oxides, carbides, etc., a cavity around the active site that may change its configuration to facilitate binding of a specific reactant to the active site and expulsion of the product, and a cavity wall that facilitates passage of the desired reactants and products from the ambient to the active site [1]. Due to the importance of active sites, there is a need to design a system where these sites are located at the surface, which enable them to freely directly interact with every substrate. Recently, Patil et al. [2] demonstrated that commercial MgO loaded with gold tested as a catalyst in the oxidation of styrene to styrene oxide with *tert*-butyl hydroperoxide (TBHP) as an oxidant, showed an excellent activity.

Here, we report the use of gold particles loaded aerogel-prepared magnesium oxide (A-MgO) as catalyst in oxidation of styrene with TBHP in liquid phase. The physicochemical properties of A-MgO have been studied previously by Klabunde et al. [3–5]. The fact that MgO was easily transformed to $Mg(OH)_2$ in the presence of water, the catalyst was then modified with alkylsilylation of *n*-octadecyltrichlorosilane (OTS) and chlorotrimethylsilane (CTMS). It is expected that the hydrophobic alkylsilyl group protects the surface and active sites from being attacked by water and side product of the oxidation reaction. A-MgO modified by attachment of titanium dioxide and surface modified with alkylsilylation of OTS and CTMS were also prepared for a comparison. The activity of catalysts was examined in the oxidation of styrene using TBHP as an oxidant.

2 Experimental

2.1 Preparation of Catalysts

2.1.1 Synthesis of Aerogel-prepared Magnesium Oxide

Synthesis of A-MgO is divided into three consecutive steps, i.e., alkoxide preparation, gel preparation and

---

H. Nur (✉) · I. I. Misnon · H. Hamdan
Ibnu Sina Institute for Fundamental Science Studies, Universiti Teknologi Malaysia (UTM), 81310 Skudai, Johor, Malaysia
e-mail: hadi@kimia.fs.utm.my; hadinur@yahoo.com
URL: http://www.hadinur.com

supercritical drying. The steps are described in the following sections.

2.1.2 *Alkoxide Preparation*

Typically, 4.8 g of magnesium ribbon (BDH Laboratory) was cleaned using acetone and cut into small pieces. The magnesium was then added to a 250 mL round bottom round flask (double neck) and fitted with a valve. The flasks were then flushed with nitrogen for 10 min in order to remove water vapor. Solution of 1 M was then prepared with addition of 206 mL methanol into the round bottom flask with continuous nitrogen flow until the reaction was complete. Nitrogen flow was needed in this reaction in order to facilitate the removal of hydrogen gas and prevent increase of pressure. In this stage, the reaction was highly exothermic. Ice bath was used to reduce heat during reaction. Finally, the reaction vessel was sealed and stored at room temperature for future use.

2.1.3 *Gel Preparation*

One Molar $Mg(OCH)_3$ solution (43 mL) in methanol was removed from the flask using a syringe and transferred to a 400 mL Teflon beaker and was mixed with 200 mL toluene. The solution was homogenized by stirring with a magnetic bar for 15 min. After that, distilled water used for hydrolysis was added dropwise to the solution. A total amount of 3.2 mL of water was slowly added at approximately 1 drop every 2–3 s with vigorous stirring. Upon addition of the water, tiny white globules were formed, which slowly disappeared turning the solution milky white. With continuous stirring, the solution became clear. The solution was covered with aluminum foil in order to prevent evaporation of the solvent and was allowed to stir overnight prior to being subjected to supercritical extraction.

2.1.4 *Supercritical Drying*

The aerogel was prepared by high temperature supercritical extraction of the gel using Parr instrument autoclave fitted with a thermocouple, a pressure gauge and a temperature controller. The autoclave with the gel was first flushed with nitrogen for 10 min. Then, it was filled with nitrogen at initial pressure of about 100 psi and sealed. The reactor was slowly heated up to 265 °C at a heating rate of about 50 °C h^{-1}. When the reactor condition reached the temperature of 265 °C, it was maintained for 1 h. After completion of the procedure, the pressure was quickly released by venting of solvent vapor and collected in the condenser. The sample was again flushed with nitrogen for 10 min at controlled pressure of 34–35 psi and allowed to cool down overnight. The resultant white powder, $Mg(OH)_2$ from the supercritical extraction was calcined in furnace at 500 °C and maintained for 2 h. The sample was cooled down to room temperature and stored in a bottle in a desiccator.

2.2 Preparation of Titanium Containing A-MgO

A series of titanium loadings were prepared (%w/w: 2.5, 5, 7.5, 10, and 15). Impregnation process was carried out according to the method reported previously [6, 7]. Typically, titanium isopropoxide $[Ti(OPr)_4$, Fluka Chemika] was impregnated from absolute ethanol solution into A-MgO by vigorous stirring at ambient condition in fume cupboard until the absolute ethanol was dried. The powder resultant was dried in an oven at 100 °C overnight. Finally, the dried sample was calcined at 500 °C for 2 h. The catalysts were labeled as xTiO$_2$/A-MgO where x refers to titanium dioxide loadings.

2.3 Preparation of Gold Containing A-MgO

Typically, 100 mL of an aqueous solution of $HAuCl_4$ with a concentration of 4.2×10^{-3} M was prepared. To the yellow clear solution, urea $[CO(NH_2)_2]$ was added to achieve a concentration of 0.42 M with the initial pH ~2. The gold concentration corresponds to a theoretically Au loading of 8 wt.% in case of a complete deposition. Then, 1 g of dried A-MgO was dispersed in the gold solution and the suspension was thermostated at 80 °C with vigorous stirring for 16 h. At the end of the reaction, the pH of the solution increased to ~10. After deposition of gold onto A-MgO, the solid was separated from the precursor solution by centrifugation (3,500 rpm for 20 min). The solid underwent washing process where it was suspended in water (100 mL) and shaken for 10 min at room temperature and centrifuged again. The washed solid was dried in an oven at 100 °C overnight and finally, the solid was calcined at 400 °C for 2 h with a heating rate of 2 °C min^{-1}.

2.4 Preparation of Alkylsilylated TiO_2/A-MgO and Au/A-MgO

Two types of alkylsilane were used: Octadecyltrichlorosilane (OTS) (Aldrich) and CTMS (Fluka). Typically, an amount of 500 μmol of alkylsilane per gram of catalyst was used for alkylsilylation. 10 cm^3 of toluene (dried over molecular sieve) was dispersed with OTS and 1 g of TiO_2/A-MgO was immersed into OTS solution.

The suspension was shaken for 15 min at room temperature and the powder was collected by centrifugation (3,000 rpm, 10 min). The catalyst powder was dried at 110 °C overnight. The catalyst was labeled as OTS-TiO_2/A-MgO. The same method was used for alkylsilylation prepared using CTMS, labeled as CTMS-TiO_2/A-MgO.

For Au/A-MgO catalyst, the modification for alkylsilylated catalyst was the same for TiO_2/A-MgO catalysts and denoted as OTS-Au/A-MgO and CTMS-Au/A-MgO.

2.5 Characterizations

The X-ray diffraction patterns were obtained using Bruker D8 Advance X-ray diffractometer using Cu K_α radiation $\lambda = 1.5418$ Å, operated at 40 kV and 40 mA. Approximately 1 g sample, in fine powder form was put into a sample holder. Then, pressed between two glass slides to get a thin layer, locked in a proper place of analyzer before it was measured. The measurement took place at room temperature in the range of $2\theta = 20°–70°$ and step interval 0.02° at a rate of 1 s per step.

Diffused reflectance ultraviolet–visible (DRUV–VIS) spectra measurement was performed on a Perkin Elmer Lambda 900 UV/VIS/NIR Spectrometer equipped with a diffused reflectance attachment and 76 mm integrating sphere. $BaSO_4$ was used as a reference. Finely ground catalyst was spread flat over the sample holder window. The holder was tightened and smoothen before placing it in the spectrometer holder. The sample was analyzed in the wavelength range of 190–700 nm.

Infrared spectra of the sample were collected on a Perkin Elmer Spectrum One spectrometer. The sample and KBr powder (1:100) was mixed using mortar and pressed into a thin pellet under vacuum (ca. 6 tons). The KBr pellet was then put into a sample holder and scanned in the wavenumber region of 4,000–400 cm^{-1}.

The specific surface areas for all samples were obtained using Quantachrome Autosorb surface area analyzer.

2.6 Catalytic Test

To examine the activity of the catalysts, oxidation of styrene was chosen as a model reaction. In the oxidation reaction, styrene (5 mmol, Fluka), anhydrous TBHP (8 mmol, Fluka), acetonitrile (5 mL) and 50 mg catalyst was put in a magnetically stirred round bottom flask (capacity 10 cm^3), under reflux (at 80 °C) for a period of 8 h. The resulting product was withdrawn and analyzed periodically with gas chromatograph (GC). Gas chromatograph-mass spectrometer (GC-MS) was also used to verify the resulting product.

3 Results and Discussion

3.1 Physical Properties of Catalysts

Figure 1 shows the XRD diffractograms of A-MgO, TiO_2/A-MgO and Au/A-MgO, respectively. The X-ray diffractogram shows that A-MgO consists of a crystalline structure with characteristic peaks at 42.9° and 62.5° due to (200) and (220) planes. After loading of Au, it clearly shows three new peaks centered at $2\theta = 38.2°$, 44.3°, and 64.5° for all samples which are ascribed to metallic gold [8]. These peaks correspond to Au(111), Au(200) and Au(220), respectively. There was no appreciable change to the X-ray pattern of TiO_2/A-MgO samples, besides a slight decrease in the intensity of peaks relative to A-MgO, confirming that A-MgO was stable towards impregnation of TiO_2. Furthermore, no TiO_2 phase was detected on all samples. This suggests that TiO_2 is either deposited in amorphous form or very well dispersed with very small crystallite size which cannot be observed by XRD [9].

DRUV–VIS spectra of A-MgO, Au/A-MgO and TiO_2/A-MgO samples are presented in Fig. 2. High surface area MgO absorbs UV light and emits luminescence, which is not observed with MgO single crystal [10]. Absorption is observed for A-MgO at wavelengths 210 nm, which is considerably lower in energy than the band at 163 nm for bulk ion pairs. The bands at 210 nm are assigned to be due to the surface O^{2-} ions of coordination number of 4. DRUV–VIS spectra for Au/A-MgO are depicted in Fig. 2b. According to the picture, Au/A-MgO exhibits a band at 537 nm, which is characteristic of the plasmon resonance of metallic metal particle [8, 11]. This brings to the assumption that gold species present in the samples consist of metallic Au. For Ti-containing catalysts, the electronic spectra of the catalysts showed absorption associated with the ligand metal charge transfer (LMCT) from the oxygen to an empty orbital of the Ti(IV) ion. Figure 2c depicts the DRUV-VIS spectra for TiO_2/A-MgO. The presence of

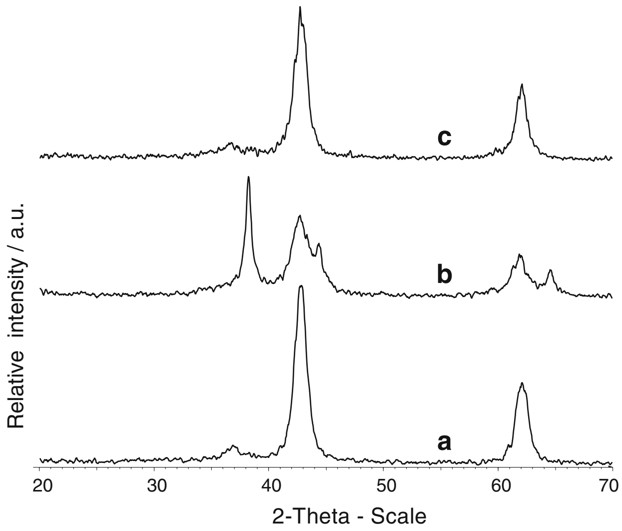


Fig. 1 X-ray diffractograms of (a) A-MgO, (b) Au/A-MgO, and (c) $10TiO_2$/A-MgO

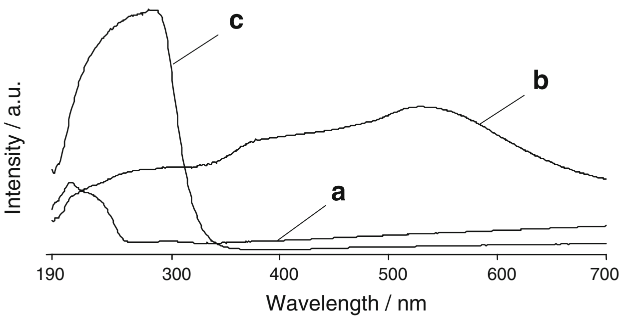


Fig. 2 DRUV–Vis spectra of (a) A-MgO, (b) Au/A-MgO, and (c) $10TiO_2$/A-MgO

octahedral titanium species in the catalysts are indicated by the appearance of intense bands centered at 280 nm.

The IR spectra of modified catalysts with OTS (OTS-TiO_2/A-MgO, OTS-Au/A-MgO) and CTMS (CTMS-TiO_2/A-MgO, CTMS-Au/A-MgO) are shown in Fig. 3a–d. The spectra demonstrate the various C–H stretching vibrations at 2,924 cm^{-1} for anti-symmetry stretching and 2851 cm^{-1} for symmetry stretching [12]. These C–H groups are from the alkyl chain of hydrophobic-inducing agent, i.e., the OTS and CTMS.

Table 1 lists the surface properties data for all the samples that have been analyzed using nitrogen adsorption analysis. Untreated A-MgO shows high surface area and pore diameter. After impregnation with TiO_2, the surface area was gradually decreased. As shown in Table 1, the surface area of catalysts containing Au was also reduced after Au deposition. However, the surface area reduction is more intense in Au deposition compared to TiO_2 impregnation. After modification by OTS and CTMS, significant decrease in the surface area was recorded. One possible reason for this is the covering effect of the internal surface area of the catalyst by alkylsilyl groups [13]. Based on the above results, it is suggested that the deposition of Au and attachment of functional groups (OTS and CTMS) occur on the surface of the A-MgO.

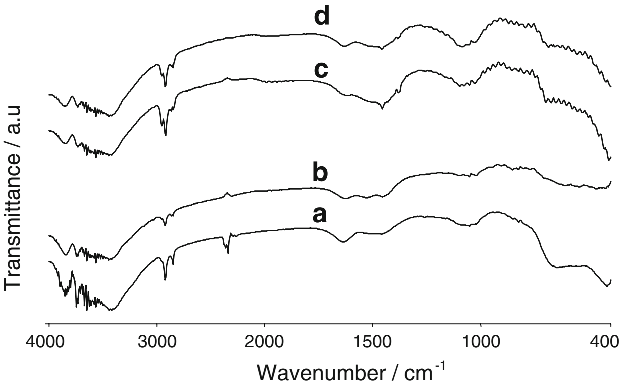


Fig. 3 FTIR spectra of (a) OTS-TiO_2/A-MgO, (b) CTMS-TiO_2/A-MgO, (c) OTS-Au/A-MgO, and (d) CTMS-Au/A-MgO

Table 1 Surface properties of aerogel prepared magnesium oxide (A-MgO) catalysts by nitrogen adsorption analysis

Sample	Surface area (m^2 g^{-1})	Total pore volume (cc g^{-1})	Average pore diameter (Å)
MgO-A	241	1.347	223
Au/MgO-A	169	1.088	257
OTS-Au/MgO-A	97	0.602	248
CTMS-Au/MgO-A	123	0.755	245
$15TiO_2$/MgO-A	187	1.423	304
OTS-$15TiO_2$/MgO-A	138	0.962	278
CTMS-$15TiO_2$/MgO-A	139	1.112	320

3.2 Catalytic Activity

3.2.1 *Catalytic Activity and Selectivity of Modified A-MgO in Oxidation of Styrene*

Figure 4 presents the catalytic activity of the catalysts in the oxidation of styrene with TBHP as the oxidant at 8 h reaction. The catalytic activity was compared at 8 h because the rate of reaction saturated after 7 h reaction. As shown in the figure, the conversion of styrene is relatively low in the absence of catalyst. TiO_2 catalyst gives a conversion of 15.9% compared to only 13.8% by A-MgO. The presence of TiO_2 increases the catalytic activity of A-MgO. It is observed that the higher is the TiO_2 loading, the higher is the conversion of styrene. This suggests that TiO_2 plays a role as catalytic active site in this reaction.

During impregnation process, the dispersion of TiO_2 in A-MgO support may lead to situation where Ti^{4+} ions are coordinated to –O–Mg entities. The presence of Ti^{4+} ions of this kind on the surface of A-MgO is believed to play an important role in the oxidation of styrene where Ti^{4+} ions is needed to activate the neighboring peroxide ligands by withdrawing the electron cloud and reducing the electron density of the oxygen atoms [9]. As another example, in the oxidation of olefin to olefin oxide using hydrogen peroxide as an oxidant, the ions (Ti^{4+}) are known to be catalytically active [6, 7, 14].

Over the catalysts, the oxidation of styrene is improved with benzaldehyde, styrene oxide and phenylacetaldehyde being the major products. As shown in Fig. 4, the selectivity towards benzaldehyde is slightly increased when the TiO_2 loading is increased. However, this trend is reversed for the selectivity of styrene oxide, where it is decreased when TiO_2 loading is increased. Selectivity towards phenylacetaldehyde on the other hand is nearly the same for all

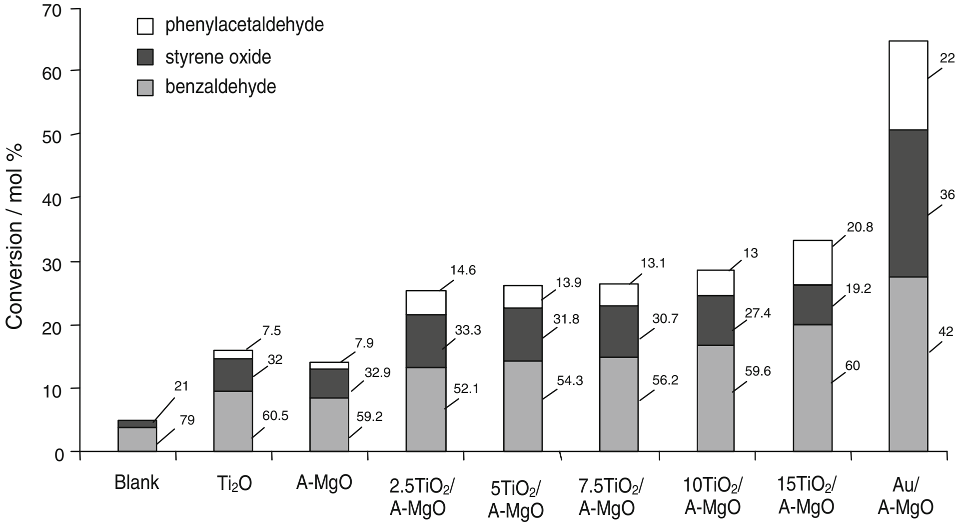


Fig. 4 Influence of titanium loading of TiO_2/A-MgO and Au/A-MgO catalysts on styrene conversion and selectivity

catalysts except for $15TiO_2$/A-MgO where it gives a slightly higher selectivity. One can see that the selectivity for benzaldehyde from styrene for all catalysts is higher than for styrene oxide and phenylacetaldehyde, particularly for the catalyst with higher TiO_2 loading ($15TiO_2$/A-MgO). One would expect that TiO_2 nano-particles are formed during modification. The TiO_2 nano-particles favored the carbon–carbon bond cleavage [15]. Therefore, with the increase in the amount of the TiO nano-particles, bond scission reaction is preferred, leading to the formation of more benzaldehyde (path I). Another way to form benzaldehyde from styrene is by oxidation reaction to form styrene oxide, which further forms benzaldehyde in the presence of peroxide (path II). A scheme of the possible pathway in styrene oxidation is depicted in Fig. 5. The two different pathways may occur in parallel; however, for this catalyst, TiO_2 promotes the oxidative cleavage of styrene [9, 16].

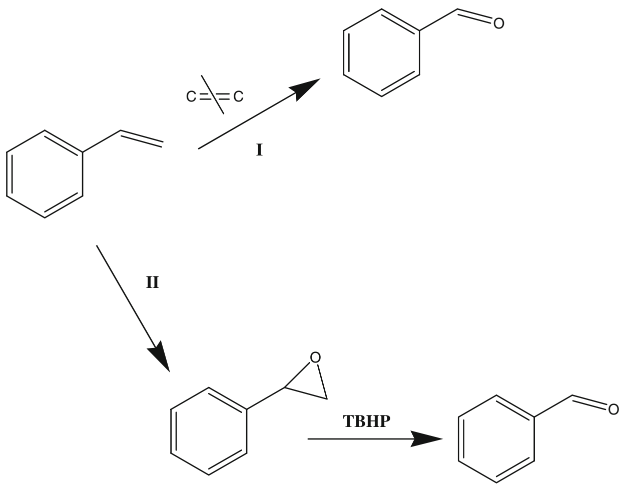


Fig. 5 Pathway in styrene oxidation reaction

As shown in Fig. 4, Au/A-MgO catalyst shows much higher activities than all TiO_2/A-MgO series. It is suggested that Au provides a better active sites for oxidation of styrene, compared to TiO_2. Au/A-MgO gives a conversion of 64.8%. Besides, Au/A-MgO increased the selectivity of styrene oxide to 36%, compared to only 19.2% with $15TiO_2$/A-MgO. It also reduced the benzaldehyde selectivity (42%) and increased selectivity towards phenylacetaldehyde (22%).

For styrene oxidation, the smaller gold particles are the most active [2]. Haruta reported that, the formation of large particles results from the fact that the interaction of $HAuCl_4$ with the support is weak, and that the chlorides present in the samples promotes the sintering of the Au particles during thermal treatment [17]. The gold particle size also depends on the calcination temperature. For example, Date and coworkers [18] showed that gold particles in Au/TiO_2 sample prepared by deposition-precipitation, grow when the calcination temperature increases. Kozlov and coworkers [19] showed that the calcination heating rates significantly alter the performances of supported gold catalysts in CO oxidation, such that low heating rates produce smaller gold particles. The calcination temperature throughout this study was set at 400 °C and the size of gold particle was big, which is ca. 15.9 nm (calculation using Scherer equation on XRD). Therefore the size of gold particles should remain the same and not be a controlling factor to the catalytic performance.

However, in this study, it can be concluded that modification of A-MgO with TiO_2 and Au helps to increase the activity of styrene oxidation. Evidently, the creation of TiO_2 and Au active sites reduce the benzaldehyde selectivity. Styrene oxide selectivity is reduced by TiO_2 and increased by Au active sites. On the other hand, TiO_2 and Au active sites increased the phenylacetaldehyde formation.

3.2.2 The Effect of Alkylsilylation

Figure 6 summarizes the conversion and selectivity of products from oxidation of styrene with anhydrous TBHP by using TiO_2 and Au series with stirring for 8 h at 80 °C. As shown in the figure, catalyzed oxidation of styrene produces benzaldehyde, styrene oxide and phenylacetaldehyde. Besides, TiO_2 and Au series enhance the styrene conversion, compared with catalysis by using A-MgO and TiO_2 alone. Furthermore, modification of support materials with hydrophobic alkylsilane groups, which is OTS and CTMS, led to a remarkable enhancement of the styrene conversion.

The improvement of catalysis result may be attributed to hydrophilicity-hydrophobicity effects. As discussed in previous reports, OTS and CTMS are effective hydrophobic inducing agents [6, 7]. During the oxidation process, decomposition of TBHP releases alcohol as a side product, known as hydrophilic substance. The hydrophilic catalyst ($15TiO_2$/A-MgO and Au/A-MgO) have an affinity to interact with alcohol molecules. If this were to take place, TiO_2 and Au active sites would be poisoned by alcohol molecules which consequently reduced the catalytic activity. So, there was a need to protect the active sites by using OTS and CTMS. Evidently, the hydrophobic carbon chain of alkylsilyl groups is proven to prevent the alcohol from deactivating TiO_2 and Au active sites. The major effect of hydrophobic behavior in the catalyst is to continuously attract more substrates and oxidizing agent towards the active sites in order to catalyze the reaction. This is because, the substrates (styrene) are hydrophobic and TBHP also have certain degree of hydrophobicity which arises from the *tert*-butyl alkyl chains. On the other hand, the hydrophilic catalyst is poisoned by the strong adsorption of water or other donating compound. Hence adsorption of hydrocarbon less occurs. Generally, as indicated in Fig. 6, modifications of TiO_2/A-MgO and Au/A-MgO catalysts with alkylsilyl groups created a high styrene conversion and their selectivities towards the formation of products are almost similar to each other.

3.2.3 Specific Catalytic Activity for A-MgO

Figures 4 and 6 represent the activity and the selectivity of the catalysts towards the oxidation of styrene. The definition of catalytic activity used in this considers the conversion of styrene per gram of catalyst. All catalysts were active towards the styrene oxidation reaction. However, one showed different performances, depending on the surface modification and the type of metals impregnated on the surface of A-MgO. As tabulated in Table 1, the surface areas of the catalysts are different. It implies that the catalytic activity site for the oxidation of styrene over the modified A-MgO depends on the surface area of catalysts. With respect to specific activity (i.e., catalytic activity based on the surface area of catalyst), alkylsilylated A-MgO having a smaller surface area showed better performance than TiO_2/A-MgO and Au/A-MgO (see Fig. 7). From these results, it can be concluded that alkylsilylated Au/A-MgO catalysts are more active compared to alkylsilylated TiO_2/A-MgO and unalkylsilylated A-MgO.

3.2.4 The Effect of Addition of Base and Drying Agent

During modification with OTS, CTMS and gold precursor, the formation of chlorine ions is favored. OTS consists of three chlorine ions while CTMS has only one chlorine ion. Although, the catalysts have been washed in order to eliminate the chlorine ions, apparently, this procedure could not completely remove the chlorine ions. Unfortunately, the presence of chlorine ions in large amount is believed to poison catalysis in many reactions [20]. In addition, if there were cations (H^+ or NH_4^+) present in the reaction system, it may bind with chlorine ions to form

Fig. 6 The effect of alkylsilane modification on A-MgO catalysts

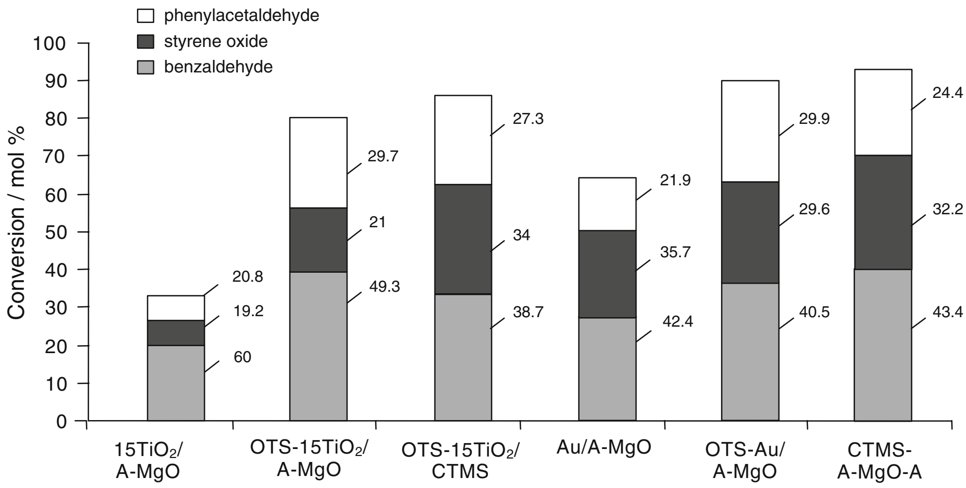
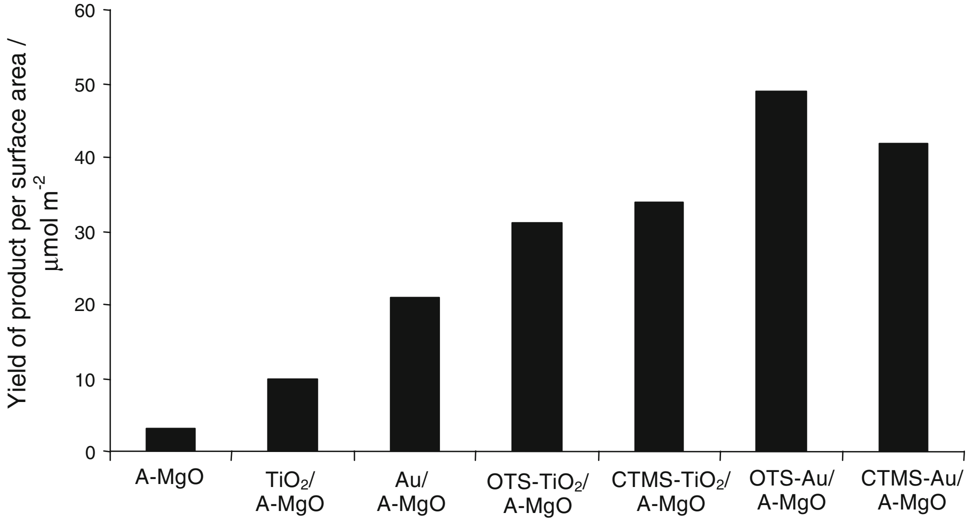


Fig. 7 Specific activity of modified A-MgO catalysts



acidic condition. Thus, a small amount of 4-nitrotoluene, an indicator used for the measurement of acid strength, was added in the catalytic system in order to deactivate the acid site. The interaction of 4-nitrotoluene with various acidic zeolites has been studied spectroscopically [21]. By considering the data tabulated in Table 2, the addition of a small amount of 4-nitrotoluene increased the activity and altered the selectivity of the reaction. OTS modified catalyst is chosen because it contains more chlorine ions than CTMS. Addition of 4-nitrotoluene gave 100% conversion for both OTS-15TiO$_2$/A-MgO and OTS-Au/A-MgO catalysts. Besides, the selectivity towards styrene oxide is highest compared to other products. Contrary to what has been reported where benzaldehyde selectivity is increased after base addition, that phenomenon obviously did not occur in this case. Evidently, phenylacetaldehyde formation is compressed as the acid is neutralized [15].

TBHP decomposition gave hydrophilic alcohol as the side product. This substance has the capabilities to reduce the catalyst performance. Although alkylsilane is introduced to reduce the deactivation effect, in this segment, drying agent was added in order to investigate the effect of water on the catalytic performance; as done by other researchers using Dean-Stark apparatus to eliminate the water from the reaction system. The presence of hydrophilic substances is also believed to promote substrate diffusion from active sites due to the different nature of hydrophilic-hydrophobic behaviors. Table 2 shows the catalyst performance after addition of drying agents. Generally, the addition of drying agent reduces the substrate diffusion, resulting in absorption of hydrophilic substance followed by a decrease in formation of phenylacetaldehyde. However, selectivity of OTS-TiO$_2$/A-MgO towards benzaldehyde and styrene oxide is increased. As discussed earlier, TiO$_2$ active sites are selective towards double bond cleavage to form benzaldehyde. On the other hand, OTS-Au/A-MgO catalyst increases the selectivity of styrene oxide and reduces the selectivity of benzaldehyde.

4 Conclusion

The catalytic potential of alkylsilylated catalysts for oxidation-catalyzed reactions is demonstrated in the liquid phase oxidation of styrene. It is observed that the catalyst is active in the oxidation of styrene with anhydrous TBHP to form benzaldehyde, styrene oxide and phenylacetaldehyde; the amount of which in decreasing order is as the following: benzaldehyde > styrene oxide > phenylacetaldehyde. The increase in oxidation activity of alkysilylated TiO$_2$/A-MgO and alkysilylated Au/A-MgO may be explained on the basis of an increase in hydrophobicity of the catalyst.

Table 2 The effect of addition of base and drying agent on product selectivity of styrene oxidation

Catalyst	Conversion (mol%)	Selectivity (%)		
		Benzaldehyde	Styrene oxide	Phenylacetaldehyde
OTS-Au/MgO-A	90.5	40.5	29.6	29.9
OTS-Au/MgO-A[a]	100	37.7	39.0	23.3
OTS-Au/MgO-A[b]	100	38.7	39.0	22.3
OTS-15TiO$_2$/MgO-A	80	49.3	21.0	29.7
OTS-15TiO$_2$/MgO-A[a]	100	37.6	44.5	17.9
OTS-15TiO$_2$/MgO-A[b]	100	60.3	27.6	12.1

[a] Addition of 4-nitrotoluene (200 µmol)

[b] Addition of molecular sieve as drying agents (100 mg)

Deactivation of the acid and reduction of water from the catalytic system improve the catalytic activity and selectivity of styrene oxidation.

Acknowledgments This research was supported by the Ministry of Science Technology and Innovation Malaysia (MOSTI), under the Sciencefund grant No. 03-01-06-SF0326 and the Ministry of Higher Education (MOHE), Malaysia under the Fundamental Research Scheme Grant Vot. 78070.

References

1. Kung HH, Kung MC (2003) Appl Catal A 246:193
2. Patil NS, Uphade BS, Jana P, Bharagava SK, Choudhary VR (2004) J Catal 223:236
3. Utamapanya S, Klabunde KJ, Schlup JR (1991) Chem Mater 3:175
4. Stark J, Klabunde KJ, Schlup JR (1993) Chem Mater 5:71
5. Klabunde KJ, Stark J, Koper O, Mohs C, Park DG, Decker S, Jiang Y, Lagadic I, Zhang D (1996) J Phys Chem 100:12142
6. Nur H, Hau NY, Misnon II, Hamdan H, Muhid MNM (2006) Mater Lett 60:2274
7. Hau NY, Misnon II, Nur H, Hamdan H, Muhid MNM (2007) J Flour Chem 128:12
8. Qian K, Jiang Z, Huang W (2006) J Mol Catal 264:26
9. Nie L, Xin KK, Li WS, Zhou XP (2007) Catal Commun 8:488
10. Hattori H (1995) Chem Rev 95:537
11. Philip D (2008) Spectro Chim Acta A 71:80
12. Dong J, Wang A, Simon-Ng KY, Mao G (2006) Thin Solid Films 515:2116
13. Mirji SA, Halligudi SB, Sawant DP, Jacob NE, Patil KR, Gaikwad AB, Pradhan SD (2006) Appl Surf Sci 252:4097
14. Laha SC, Kumar R (2001) J Catal 204:64
15. Zhuang J, Ma D, Yan Z, Liu X, Han X, Bao X, Zhang Y, Guo X, Wang X (2004) Appl Catal A 258:1
16. Nur H, Hayati F, Hamdan H (2007) Catal Commun 8:2007–2011
17. Haruta M (2002) Cattech 6:102
18. Date M, Ichihashi Y, Yamashita T, Chiorino A, Boccuzzi F, Haruta M (2002) Catal Today 72:89
19. Kozlov AI, Kozlova AP, Asakura K, Matsui Y, Kogure T, Shido T, Iwasawa Y (2000) J Catal 196:56
20. Zanella R, Giorgio S, Henry CR, Louis C (2002) J Phys Chem B 106:7634
21. Umansky B, Englehardt J, Hall WK (1991) J Catal 127:128