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Rimeh Ismail, Jihene Arfaoui, Zouhaier Ksibi, Abdelhamid Ghorbel, Gerard Delahay. Ag/ZrO2 and Ag/Fe–ZrO2 catalysts for the low temperature total oxidation of toluene in the presence of water vapor. Transition Metal Chemistry, 2020, 45 (7), pp.501-509. 10.1007/s11243-020-00402-y . hal-02954522

HAL Id: hal-02954522
https://hal.science/hal-02954522

Submitted on 26 Nov 2020

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Ag/ZrO_2 and $Ag/Fe–ZrO_2$ catalysts for the low temperature total oxidation of toluene in the presence of water vapor

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Abstract

New mesoporous and well-structured Ag-based catalysts (**x**Ag/ZrO_2 and **x**$Ag/Fe–ZrO_2$ with **x** = 2 wt%) have been investigatedin the total oxidation of toluene with water vapor. The results showed that the catalytic activity of systems increases, in the 350–550 °C temperature range, following this order: ZrO_2< $Fe–ZrO_2$ < Ag/ZrO_2 ≈ $Ag/Fe–ZrO_2$. Nevertheless, it was revealed that $Ag/Fe–ZrO_2$ is the most active solid for the low temperature total oxidation of toluene (< 350 °C). The highest low temperature activity of this new catalyst can be related to the presence of more reactive surface oxygen, new active redox and acidic sites at $Ag/Fe–ZrO_2$ surface, most probably generated by the simultaneous presence of Ag and Fe species.

Keywords Aerogel supports · Ag/ZrO_2 · $Ag/Fe–ZrO_2$ · Toluene oxidation · Water vapor

Introduction

Volatile organic compounds (VOCs), like aldehydes, alcohols, ketones, aromatic, halogenated and sulfur containing compounds, which can be emitted from automobile industries, petroleum refineries, textile manufacturers, chemical industries, solvents processes, fuel combustions, pharmaceuticalplants, decomposition in the biosphere and biomass, etc., remain one of the major sources of air pollution [1, 2].Therefore, it is necessary to control and reduce the emissions of these compounds. Destructive methods (catalysis oxidation and biofiltration) and recovery methods (condensation, adsorption, absorption, and membrane separation) have been developed for VOCs removal [3]. Among the available technologies, the catalytic oxidation is most popular and effective because of its versatility of eliminating a range of organic pollutants under mild operating conditions [1]. On the other hand, this technique is relatively more environmentally friendly due to its low temperature operation and the formation of less dioxins and noxious products [1]. Divers catalytic systems containing noble or transition metals (e.g., Pd, Pt, Mn, Fe, etc.) have been investigated and demonstrated a high catalytic performances for the destruction of various VOCs (e.g., propane, carbon monoxide, methane, toluene, etc.) [1–11].

In recent years, silver- based materials have been extensively investigated and recognized as suitable catalysts for the low temperatures oxidation of VOCs [6, 12]. These catalysts bring about a special interest due to a number of valuable features of Ag including its ability to activate molecular oxygen, homogeneous distribution of metal particles/clustersover various supports [e.g., TiO2, ZrO2, SiO2, CeO2, MnxOy, Si3N4, Fe2O3, etc.] and a synergistic action

with a large number of promoters and modifiers (noble and transition metals, halogen-containing compounds, etc.) [12]. Recently, the activity exploration focused on various Ag supported catalysts has attracted increasing interest in the oxidation of toluene which is known as one of the most targeted components among VOCs [6, 13, 14]. In this framework, Qu et al. [13] have showed that the addition of Ag greatly enhances the reactivity of Mn/SBA-15 for toluene oxidation. The authors have reported that the strong interactions between Ag and Mn species promotes both the reducibility of catalysts and the formation of abundant active lattice oxygen, thus increasing the catalytic activity. On the other hand, Zhang et al. [6] have studied the effect of Ag loading on the structure and performance of the Ag nanoparticles supported on UiO-66 catalysts for toluene oxidation. They have concluded that the increase of the Ag amount affects the UiO-66 structure and the 10Ag-U catalyst showed excellent catalytic performance due to higher lattice oxygen and surface Ag^0 content. Furthermore, Lu et al. [14] have studied the removal of toluene by plasma-catalytic oxidation (non-thermal plasma (NTP)) over Ag-modified FeO**x** / SBA-15 solids. Their results revealed that the incorporation of FeO**x** and AgO**x** significantly enhances the toluene removal efficiency and CO2 selectivity of the NTP systems. The authors have reported that the silver particles tended tocover the catalyst surface in the form of Ag and Ag2 O, which improves the dispersion of FeO**x** and lengthened the residual time of toluene in the NTP-catalytic system, there by promoting the reaction of toluene with oxygen on the catalyst surface increasing toluene oxidation and catalyst stability.

To the best of our knowledge, up to date, no studies related to the total oxidation of toluene (C6 H5 –CH3) over Ag and Fe supported on ZrO2 solid, has been described in the literature. Therefore, we reported in this work the preparation and characterization of new Ag/ZrO2 , Fe–ZrO2 , and Ag/Fe–ZrO2 catalytic systems for the total oxidation of toluene in the presence of water vapor. ZrO2 and Fe–ZrO2 catalytic supports were prepared by the one step sol–gel procedure which represent one of the low cost techniques that can easily be applied to large area deposition [15 , 16]. This method allows the facile synthesis of stable metal oxides with better purity and homogeneity at ambient conditions [15]. It gives high chemical homogeneity, provides good dispersion nanoparticles, well-controlled composition, and useful for the applications where large area coatings are required [15]. Silver was deposited at ZrO2 and Fe–ZrO2 surface by the impregnation method. The mutual relationship between the physicochemical properties and catalytic activity in the toluene oxidation of Ag/ZrO2 , Fe–ZrO2 , and Ag/Fe–ZrO2 samples has been discussed.

Experimental

Catalysts preparation

ZrO2 and Fe–ZrO2 aerogel supports were prepared by the sol–gel method using a similar procedure to that reported in our previous work for TiO2 [17]. In fact, the requisite quantities of Zirconium (IV) butoxide (Zr(OC4H9)4, Sigmaaldrich, Purity (**p**) = 80 %) and Ethyl acetoacetate (C6H10O3, Fluka, Purity (**p**) > 99.5% Sigma-aldrich, **p** = 80%) were dissolved in anhydrous ethanol (C2H6O, Aldrich, **p** = 99.8%) and mixed under stirring at room temperature. Then, a dilute solution of HNO3 (0.1 M) was gradually added with a suitable molar ratio nH2O/nZr = 10 to control hydrolysis. Finally, the mixture was kept under continuous stirring at
room temperature until gelification. For Fe–ZrO2 solid, an appropriate amount of iron(III) nitrate (Fe(NO3)3,6H2O), Sigma-aldrich, p ≥ 99.95%) was introduced in the mixture before hydrolysis. The resulting gel were dried in autoclave (at T = 243 °C and P = 63 bar) to form the aerogel supports (ZrO2 , Fe–ZrO2) which were calcined at 550 °C for 3 h under O2 flow (30 cm3 /min). The Ag/ZrO2 and Ag/Fe–ZrO2 catalysts, containing a theoretical loading 2 wt% of Ag, were prepared by the

standard impregnation method. Hence, the required quantity of $AgNO_3$ (Sigma-Aldrich, p ≥ 99.0% was dissolved in water (H_2O) and the obtained clear solution was mixed under stirring with ZrO_2 or Fe–ZrO_2 powder to obtain Ag/ZrO_2 or Ag/Fe–ZrO_2 sample which was dried at 100 °C for 12 h and subsequently calcined at 550 °C for 3 h under O_2 flow (30 cm^3 /min).

Catalytic tests

The toluene oxidation tests were carried out in a fixed bed reactor using 20 mg of catalyst which was packed between the layers of a quartz wool. The catalyst bed temperature was monitored and controlled by a temperature controller equipped with two thermocouples inserted at the catalyst reactor bed and furnace. The toluene concentration was adjusted by passing an appropriate He flow through a saturator, kept at 0.7 °C, containing toluene. A total flow rate of 50 cm^3 /min gas mixture containing 0.1 vol% toluene (purity; p > 99.7%), 3.4 vol% H_2O (p > 99.5%), 8 vol% O_2 (p > 99.9%) and balance with He (p > 99.9%) were used. The equivalent gas hourly space velocity (GHSV) obtained was of 60,000 h^{-1} . The catalytic activity was evaluated in terms of toluene conversion to CO_2 ,in the 200–550 °C temperature range, with a ramp of 6 °C/min. Exit gases were monitored continuously by sampling on line to a quadrupole mass gas spectrometer Pfeiffer Vacuum equipped with Faraday and SEM detectors (0–200 amu).

The toluene conversion (%) is expressed as:

$$X_{tot}(\%) = \frac{[\text{Toluene}]_{in} - [\text{Toluene}]_{out}}{[\text{Toluene}]_{in}} * 100,$$

where $[Toluene]_{in}$ and $[Toluene]_{out}$ are the inlet and outlet concentrations of toluene, respectively.

The concentration of CO_2 (ppm) was calculated as given below:

$$[CO_2]\,(\text{ppm}) = \frac{[I]_t - [I]_0}{[I]_{7000}} * 7000,$$

where $[I]_0$, $[I]_{7000}$ and $[I]_t$ are the intensity of CO_2 signals at inlet, outlet and at any reaction temperature (t), respectively. 7000 ppm is the inlet concentration of CO_2 .

Catalysts characterization

Textural properties of the samples were determined by N2-physisorpton at 77 K using a Micromeritics ASAP 2000 instrument. The samples were previously degassed for 5 h at 250 °C. Structural features were investigated by a Brüker AXS D8 diffractometer with CuKα radiation (λ = 1.5406 Å) and the spectra were collected using a step size of 0.02° in the range 20° and 70°. JCPDS powder diffraction files were used for phase identification. Redox behaviors of solids were analyzed by temperature-programmed reduction technique (H2-TPR) with an AUTOCHEM 2910 (Micromeritics). Prior to H2-TPR measurements, the sample (80 mg) was pre-treated under 5 vol% O2 in He (30 cm3/min) at 550 °C (ramp 10 °C min−1) for 30 min. After being cooled down to 50 °C in the same atmosphere, the sample was flushed with He (30 cm3/min) then exposed to a flow containing
5 vol% H2 in Ar (30 cm3/min) and heated between 50 °C and 800 °C with a heating rate of 10°C/min. Acidity of samples was examined by NH3-TPD using an AUTOCHEM 2910 (Micromeritics). The solid was pre-treated at 550 °C (ramp 10 °C/min) during 2 h, under air flow (30 cm3/min). Then, it was exposed to 5 vol% NH3 in He (flow rate = 30 cm3/min) for 30 min to be flushed after with He (30 cm3/
min) during 30 min to remove the physisorbed ammonia. Finally, the NH3 desorption was

done in helium flow (30 cm3/ min) from 100 to 550 °C (the heating rate was 10 °C/min).

Results and discussion

The samples (ZrO2, Fe–ZrO2, Ag/ZrO2 and Ag/Fe–ZrO2) were tested in the toluene (C6H5–CH3) oxidation in the presence of water vapor (3.4 vol%). The collected results (C6H5–CH3 conversion and CO2 concentration as temperature reaction function) are shown in Figs. 1A, B.

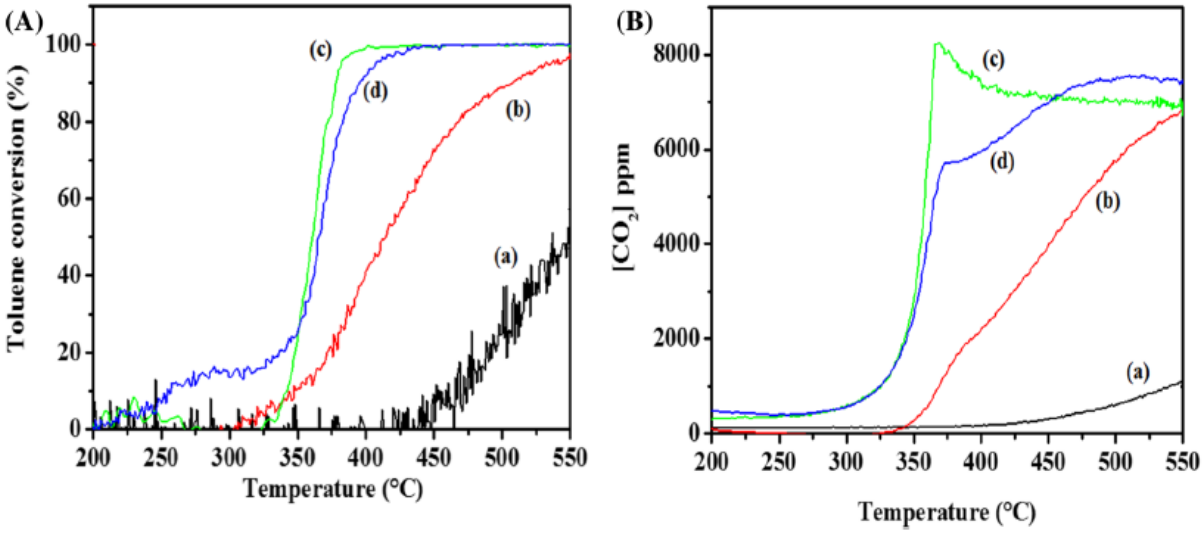


Fig. 1 **A** Toluene conversion and **B** CO2 concentration (ppm) over (a) ZrO2, (b) Fe–ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe–ZrO2 samples

The temperatures corresponding to 10%, 50%, and 90% of toluene conversions (denoted as T10, T50, and T90) are used to evaluate and compare the catalytic activities of solids.

It is well established that the mechanism of VOCs oxidation over transition metal oxides follows a Mars–Van Krevelen mechanism, which is known as redox mechanism, involving two successive steps in terms of the cyclic reaction. In the first step, oxygen vacancies on the catalyst surface were reduced as they react with the organic molecules. In the second step, the pre-formed reduced site regenerated immediately through the consumption of gaseous oxygen or the transfer of oxygen atoms from the bulk to the surface [18–20]. Therefore, the oxygen carrier capacities (adsorbed and lattice oxygen species) and reducibility of catalyst determined its reactivity in the oxidation of VOCs [18–21]. Noting that the surface acidity of catalyst also play a key role in the oxidation reaction since it could facilitate both the sorption of VOC at the catalyst surface and the breaking of carbon–carbon bonds which promote the CO2 production [22].

As it can be seen in Fig. 1, the ZrO2 support exhibits the lowest C6H5–CH3 conversions to CO2 (a maximum of 50% at 550 °C) due to the low reactivity of its acidic sites in this reaction and the absence of redox ones. However, the addition of iron or silver species significantly enhances the activity for the toluene oxidation. Higher C6H5–CH3 conversions to CO2 are obtained in the 350–550 °C temperature range over Ag/ZrO2 (near 100%) compared to that observed in the case of the aerogel Fe–ZrO2 (a maximum of 95% at 550 °C). This result demonstrates that the actives sites (most probably acidic and redox

ones) created by silver species are more reactive in the toluene oxidation than those generated by iron ones. The temperatures at which toluene conversion reaches 10%, 50%, and 90%, over Fe–ZrO2, Ag/ZrO2 and Ag/Fe–ZrO2 catalysts, are indicated in Table 1.

The catalytic activity test demonstrates that Ag/ZrO2 and Ag/Fe–ZrO2, which were found more active than Fe–ZrO2 (Fig. 1A, B), exhibit approximately a similar oxidation activity at high temperatures (100% of toluene conversion for T > 400 °C). However, the simultaneous presence of Ag and Fe leads to the most active Ag/Fe–ZrO2 catalyst in the low temperatures range, therefore, T10 are 349 °C, 341 °C and 250 °C over Fe–ZrO2, Ag/ZrO2 and Ag/Fe–ZrO2, respectively. Based on this result, we can suggest the existence of Ag-Fe interactions, which promote the dispersion of silver and iron active species, facilitates the mobility and activation of oxygen, and enhance the reactivity of acidic and redox sites, thereby increasing the low temperature activity of Ag/Fe–ZrO2 catalyst in the total oxidation of toluene. This is in perfect agreement with the result already obtained by Lu et al. [14] for the non-thermal plasma oxidation of toluene. In fact, the authors have demonstrated that 2%FeOx–1%AgOy/SBA-15 catalyst exhibits higher stability, toluene removal efficiency and CO2 selectivity than 3%FeOx/SBA-15 system. Their results revealed that silver, existing in the form of Ag particles and Ag2O on the catalyst surface, adsorb toluene and promote the production of hydroxygroups, thus improving the reductive activity of FeOx.
The authors have concluded that the interaction between Ag and Fe species could increases the catalyst activity and consequently improves the performance of the plasma-catalytic degradation of toluene. Meanwhile, Scirè et al. [23] reported that Ag could weaken the Fe–O bond, increase the mobility of the lattice oxygen, and thus improve the VOCs oxidation. By contrast, Rioseco et al. [24] have concluded that no increase in the catalytic activity for the complete combustion of toluene was observed upon low Ag substitution in LaFeO3 perovskite.

Table 1 Conversions of toluene at different temperatures over thecatalysts

Catalyst	*T*10 (°C)	*T*50 (°C)	*T*90 (°C)
ZrO_2	469	550	–
Fe–ZrO_2	349	414	506
Ag/ZrO_2	341	360	377
Ag/Fe–ZrO_2	~ 250	365	394

Table 2 Catalytic activity of silver-based catalysts in thetoluene oxidation

Catalyst	*T*10 (°C)	*T*50 (°C)	*T*90 (°C)	Reference	Catalytic conditions
Ag/Fe–ZrO_2	~ 250	365	394	This work	GHSV = 60.000 h^{-1}, m = 20 mg With 3.4 vol% H_2O
Ag/γAl_2O_3	~ 330	~ 360	~ 380	[25]	GHSV = 15,000 h^{-1}, m = 200 mg Without H_2O
Ag/Cu/Al_2O_3	~ 260	~ 280	~ 330	[25]	GHSV = 15,000 h^{-1}, m = 200 mg Without H_2O

With: GHSV = gas hourly space velocity and m = mass of catalyst

It is worthy to note that, the efficiency obtained over the new Ag/Fe–ZrO2 catalyst in the

low temperature total oxidation of toluene (particularly T10) is better compared to some literature data (Table 2) since the reaction was performed inthis work with water vapor (3.4 vol%), at higher gas hourly space velocity (GHSV = 60,000 h−1) and with a low amount of catalyst (m = 20 mg).

The results of the N2 physisorption analysis, given in Table 3, reveal that all the samples exhibit after calcination at 550 °C developed texture (high surface area (SBET ≥ 76m2/g) and total pore volume (VPT ≥ 0.24 cm3/g)) demonstrating a good thermal stability of catalysts. Nevertheless, it should be mentioned that a slight decrease of the SBET and VPT accompanied by an increase of the pore diameter of ZrO2 is observed after Fe and/or Ag addition. This might be attributed, on one hand, to the partial obstruction of some pores of solids by the supported species [26], and on the other hand, to the increase of the crystallite size of zirconia after its modification by iron and/or silver species [27]. Similar results have been obtained by Dastan et al. [27] upon heat treatment of TiO2 NP's system. The authors have explained the decrease of the surface area and pore volume of this later solid, with the increase of its pore diameter, upon heat treatment, to the increase in the crystallite size and the collapse of pore structure of TiO2 NP's at higher annealing temperature.

Table 3 Textural properties of samples

Catalyst	Surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore diameter (Φpore, Å)
ZrO_2	107	0.33	88
$Fe–ZrO_2$	93	0.28	92
Ag/ZrO_2	76	0.24	97
$Ag/Fe–ZrO_2$	84	0.26	116

Based on the N2 adsorption–desorption isotherms and BJH pore size distribution curves of solids (shown in Figs. 2 and 3, respectively) it can be concluded that all the samples are mesoporous systems (isotherms type IV according to the IUPAC classification) with monomodal (case of ZrO2 and Ag/ZrO2: hysteresis loops type H2) or binodal (for Fe–ZrO2 and Ag/Fe–ZrO2: hysteresis loops type H2) porous distribution.
It should be also mentioned that the increase of zirconia pores size after the iron and/or silver addition could indicate the existence of divers interactions between ZrO2 and the supported species which probably affects the texture of solids as previously suggested for various aerogels catalysts [17].

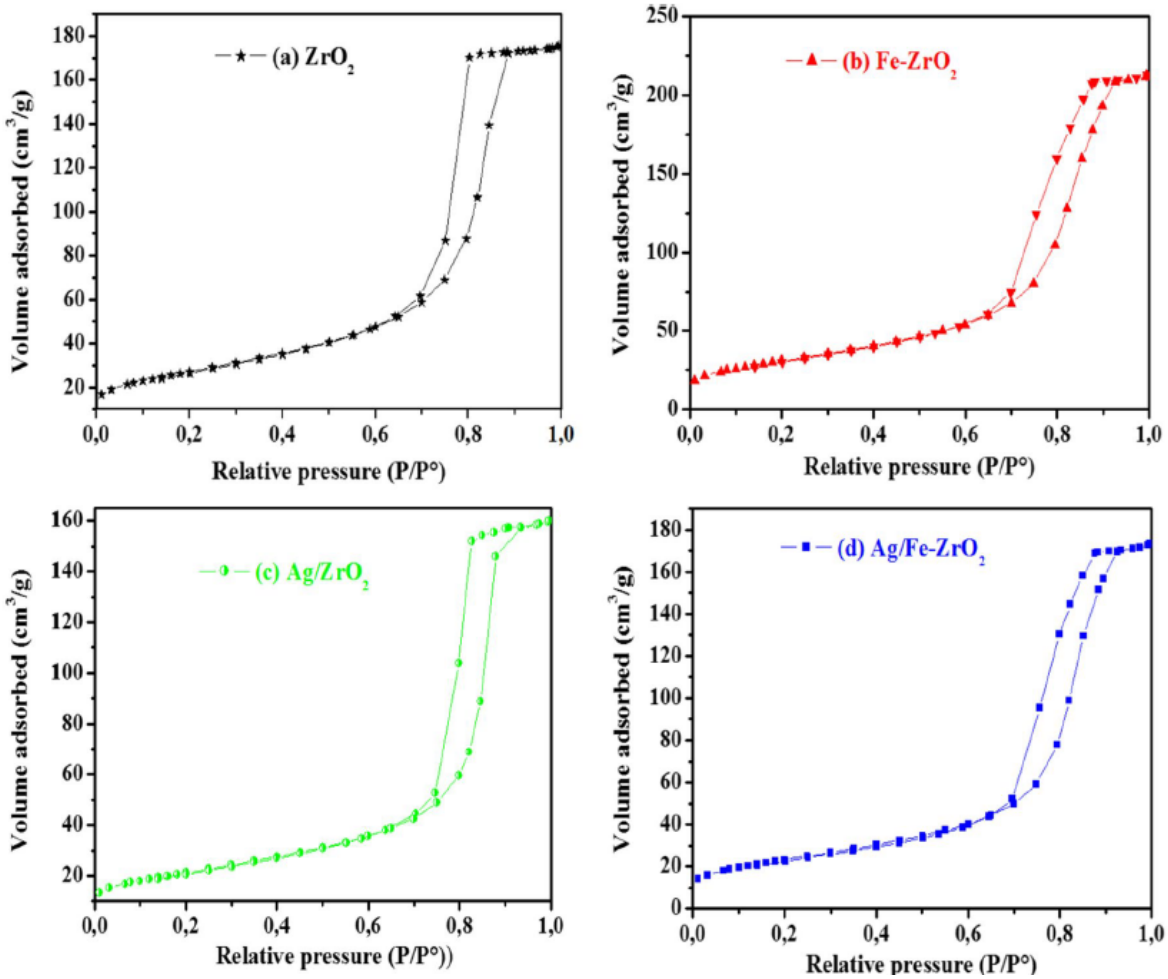
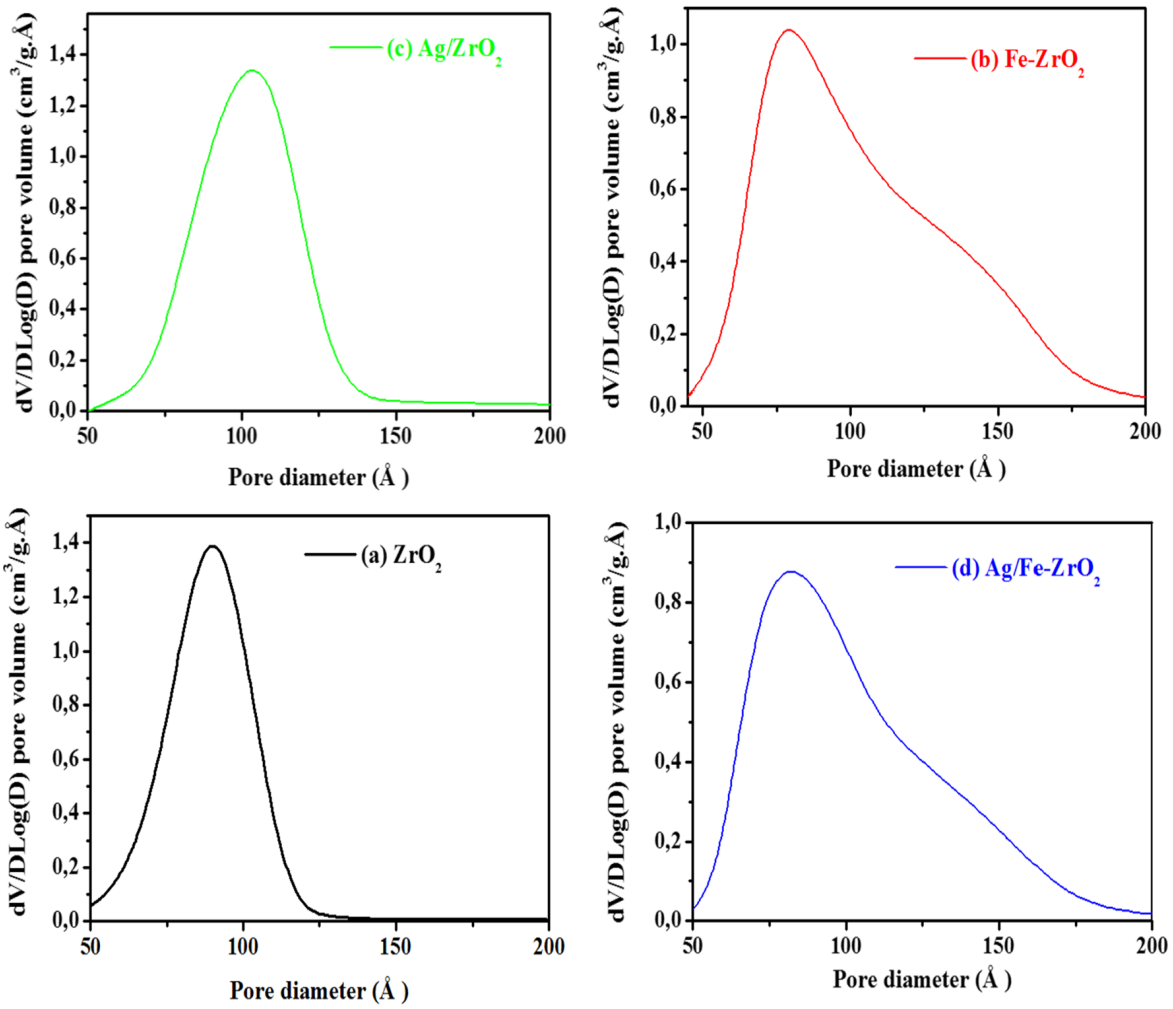


Fig. 2 N2 adsorption–desorption isotherms of (a) ZrO2, (b) Fe-ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe-ZrO2 samples



` Fig. 3 BJH pore size distribution curves of (a) ZrO2, (b) Fe-ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe-ZrO2 samples

Figure 4 shows the XRD diffractograms of samples. It is clearly seen that all the solids develop a high crystalline structure of zirconia. Hence, two major diffraction lines corresponding to the monoclinic (m-ZrO2) and tetragonal phases (t-ZrO2) of ZrO2 are detected, on the diffractograms of ZrO2 and Ag/ZrO2 solids, at 2θ = 24.38° (110), 28.26° (− 111), 31.56° (111), 41.07° (102), 45.21° (− 202), 54.09° (300), 55.60 (130), 65.68° (320) [PDF 89-9066] and 2θ = 30.33° (101), 34.67° (002), 50.37° (112), 60.09° (211), 62.57° (202) [PDF 79-1769], respectively. Nevertheless, only the typical peaks of the t-ZrO2 phase are observed in the XRD patterns of Fe–ZrO2 and Ag/Fe–ZrO2 catalysts. This suggests the stabilization of the tetragonal phase of zirconia by its interactions with the supported Ag and Fe species.

Markedly, no reflections associated with metallic iron or its oxide phases are found. This implies that Fe species are highly dispersed on ZrO2 surface. However, a weak diffraction line corresponding to the metallic silver was detected at around 2θ = 38.1° (111) for Ag/ZrO2 sample revealing the presence of a trace amount of metallic Ag (PDF 65-2871). This result suggests the existence of Ag–Fe interactions which contribute to the high dispersion of silver species at the catalyst surface.

The crystallites size of pure and modified zirconia is calculated using the Scherrer's formula [28–30] and the obtained values are 10.6, 18, 11.8, and 19.8 nm for ZrO2, Fe–ZrO2, Ag/ZrO2, and Ag/Fe–ZrO2, respectively.

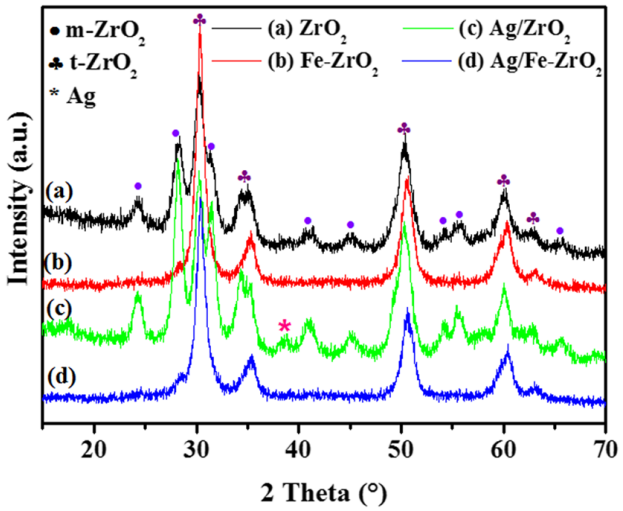


Fig. 4 XRD patterns of (a) ZrO2, (b) Fe–ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe–ZrO2 samples

Based on this result, it can be concluded that all the samples are characterized by a nanometer size and the addition of Ag and/or Fe contributes to the crystal growth of zirconia.

H2-temperature-programmed reduction (H2-TPR analysis) was performed to study the redox properties of materials and the results are illustrated in Fig. 5.

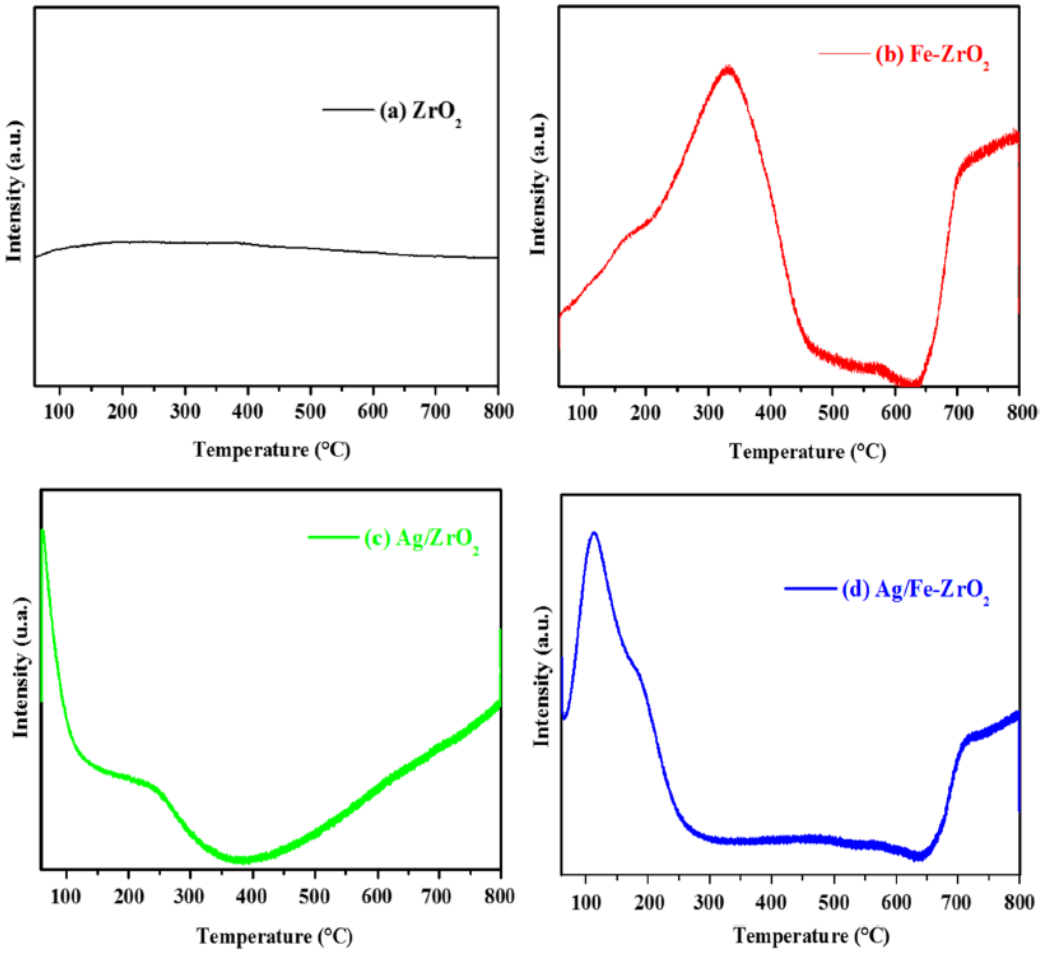


Fig. 5 H2-TPR of (a) ZrO2, (b) Fe–ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe–ZrO2 samples

For the ZrO2 solid, no H2 consumption peaks are detected indicating that Zr4+ is a non-reducible cation [31]. For the aerogel Fe–ZrO2, two main H2-consumption peaks are appeared in the experimental temperature range; the peak at low temperatures (centered at T ≈ 150 °C) could be attributed to the reduction of Fe2O3 to Fe3O4 and the other one, observed at higher temperatures (centered at T ≈ 350 °C), can be associated with the Fe2O3 → Fe3O4 → Fe^0 transformations [32]. Similarly, a new H2- consumption peaks are observed at low temperatures (< 300 °C) in the H2-TPR profile of Ag/ZrO2 and Ag/Fe–ZrO2 catalyst and can be ascribed to the reduction of both silver and iron species [33, 34]. It is noteworthy that after the addition of Ag to Fe–ZrO2 solid, the reduction peaks of Fe species was shifted to the lower temperatures indicating an enhancement of the iron species reducibility, must probably due to their interactions with silver which is consistent with the XRD results. Similar results, suggesting the improvement of FeOx reducibility by silver addition, have been previously obtained by Lu et al. [14] for 2%FeOx–1%AgOy/SBA-15 system.

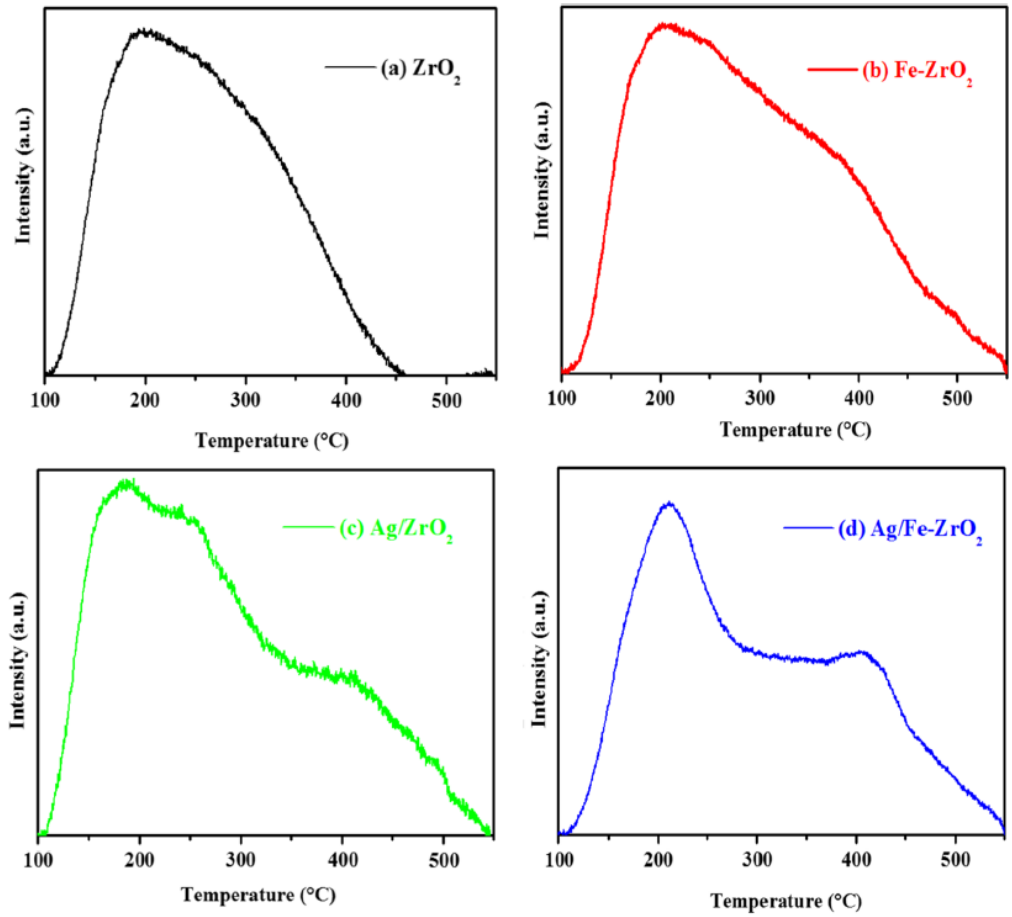


Fig. 6 NH3-TPD of (a) ZrO2, (b) Fe–ZrO2, (c) Ag/ZrO2, and (d) Ag/Fe–ZrO2 samples

The total acidity of solids was evaluated using the temperature-programmed desorption of ammonia (NH3-TPD) and the results are presented in Fig. 6. The ZrO2 and Fe–ZrO2 aerogel supports show a broad NH3 desorption peak between 100 and 400 °C corresponding to NH3 desorbed from weak and medium strong acid sites [35]. The intensity of this former peak is higher for the solid containing Fe compared to that characterizing pure

zirconia. This indicates that the presence of iron increases the total acidity of the support and contributes to the creation of new strong acid sites (NH3 desorbed at T > 400 °C). The addition of silver affects the total acidity of ZrO2 and Fe–ZrO2 solids and leads also to the creating of new strong acid sites (desorption of NH3 at T > 400 °C), especially, at Ag/Fe–ZrO2 surface.

Conclusions

In this work, a highly crystalline and mesoporous ZrO2, Fe–ZrO2, Ag/ZrO2, and Ag/Fe–ZrO2 catalytic systems were prepared for the total oxidation of toluene in the presence of water vapor. It was shown that the presence of Fe or Ag stabilizes the tetragonal phase of ZrO2 and leads to the creation of new redox and strong acid sites at zirconia surface. On the other hand, it was revealed that the coexistence of Ag and Fe species promotes both the acidity and reducibility of catalysts and contributes to the formation of more reactive oxygen which is beneficial for the low temperature toluene oxidation. Ag/Fe–ZrO2 was found to be the most active catalyst in the low temperature total oxidation of toluene (T10 = 250 °C) and exhibits approximately similar catalytic performances to that obtained over Ag/ZrO2 at higher temperatures (350–550 °C). Interestingly, 100% toluene conversion to essentially CO2 is obtained in 400–550 °C temperature range over the new Ag/Fe–ZrO2 catalyst.

Compliance with ethical standards

Conflict of interest The authors declare that there is no conflict of interest regarding the publication of this article.

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