New CeO2–TiO2, WO3–TiO2 and WO3–CeO2–TiO2 mesoporous aerogel catalysts for the low temperature selective catalytic reduction of NO by NH3

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New CeO_2-TiO_2, WO_3-TiO_2 and WO_3-CeO_2-TiO_2 mesoporous aerogel catalysts for the low temperature selective catalytic reduction of NO by NH_3

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Abstract

New mesoporous and well-structured aerogel catalysts (CeO_2-TiO_2, WO_3-TiO_2 and WO_3-CeO_2-TiO_2) were elaborated via the sol gel method, characterized by means of various techniques (XRD; N_2-Physisorption at 77 K; NH_3-TPD; H_2-TPR; DRUV-Vis spectroscopy) and evaluated in the selective catalytic reduction (SCR) of NO by NH_3. The results reveal that all the aerogel catalysts develop essentially the diffraction peaks of TiO_2 anatase phase and are classified as mesoporous materials with a high surface area ($70 < S_{BET} < 106$ m^2/g), large porosity ($0.27 < V_{PT} < 0.46$ cm^3/g) and nanometer size of crystallites (8-15 nmm). The addition of Ce and/or W influences differently the structure, texture, crystallites size, surface oxygen concentration, total acidity and redox ability of aerogel samples and clearly affects their NO-SCR activity which follows this order: TiO_2 < WO_3-TiO_2 < CeO_2-TiO_2 < WO_3-CeO_2-TiO_2. It was also found that cerium species are more active in the low temperature NO-SCR reaction than tungsten ones (NO conversions obtained at 300 °C using CeO_2-TiO_2 and WO_3-TiO_2 were 75 % and 0 %, respectively). On the other hand, it was suggested that the interactions between Ce and W species play a key role in improving the reactivity of WO_3-CeO_2-TiO_2 catalyst in the SCR of NO by NH_3. Interestingly, the NO conversion into N_2 reaches 85 % at 300 °C and exceeds 90 % between 320 and 400 °C over this novel meso-structured aerogel catalyst.

Key words: Sol Gel method, supercritical drying, mesoporous materials, New WO_3-CeO_2-TiO_2 aerogel catalyst, W↔Ce interactions, NO-SCR by NH_3

1. Introduction

Nitrogen oxides (NO_x), emitted from the combustion of fuels in stationary and mobile processes, are among the main atmospheric pollutants that cause a variety of harmful environmental and human heath effects [1,2]. In order to meet the stringent environmental regulations, research in the field of NO_x abatement has grown significantly in the past two decades. Up to date, the selective catalytic reduction using NH_3 (NH_3-SCR) as reductant is still the most powerful method for NO_x removing from stationary source and diesel engines and the commercial catalysts for this reaction are V_2O_5-WO_3/TiO_2 or V_2O_5-MoO_3/TiO_2 [3]. Although such catalysts showed sufficient activity and stability in the 300-400°C temperature range, vanadium-based systems are still not satisfactory due to a series of problems such as the toxicity of V_2O_5, low N_2 selectivity at high temperature and narrow operation temperature window [4]. Therefore, there is still a big challenge to develop a novel vanadium-free and environmentally friendly catalysts for the SCR process [5].

In recent years, the research of cerium-based catalysts in NH_3-SCR reaction has attracted great attention due to the excellent redox performance, remarkable oxygen storage capacity, low cost and non-toxicity of cerium [6,7]. A variety of Ce based catalysts, such as : CeO_2/TiO_2 [8,9], CeO_2/ZrO_2 [10], CeO_2/Al_2O_3 [11], CeO_2-Nb_2O_5 [12] CeO_2/ MnOx [13] CeO_2/WO_3 [14], Ce-titanium nanotubes [15], CeZSM5 [16], Ce-activated carbon fiber [17], Ce-carbon nanotubes [18] and Mn-Ce-Ti [19] have been developed for the NH_3-SCR reaction. Among them, CeO_2/TiO_2 systems have been widely applied for industrial denitrification due to its wide active window and environmental friendliness [20]. On the other hand, it was reported that ceria based catalysts containing acidic components (such as SO_4^{2-} and WO_3) show high NH_3-SCR activity and N_2 selectivity in a wide temperature window [4]. Recentlty, more and more attention has been devoted to W-Ce-Ti SCR catalysts [4,20-25]. The studies demonstrated that the addition of WO_3 can significantly enhance the acidic and redox properties of CeO_2/TiO_2 catalysts, thereby improve their NH_3-SCR performance.

Chen et al. [21] found that the standard SCR reaction ($4\ NH_3 + 4\ NO + O_2 \rightarrow 4\ N_2 + 6\ H_2O$) was enhanced over CeO_2/TiO_2 catalyst after WO_3 addition. In fact, it was shown that CeWTi solid provides more adsorbed NOx and NH_3 species and simultaneously increases the reactivity of both species resulting in the improvement of the SCR activity. Furthermore, it was reported that the strong interaction between Ce and W results in more Ce^{3+}, which creates charge imbalance facilitating more chemisorbed oxygen on the surface of catalysts [26]. According to Tronconi et al. [27], the chemisorbed oxygen is considered as the most active oxygen for NO_2 formation in the ‘‘fast SCR’’ reaction ($2\ NH_3 + NO + NO_2 \rightarrow 2\ N_2 + 3\ H_2O$) for which the reaction rate is much faster than that in standard NH_3-SCR reaction. Meanwhile, W. Xie et al. [25] concluded that the synergistic effects of CeO_2 and WO_3

produce more active oxygen species, increase the number of acidic sites and promote the SCR activity of many Ce-W based catalysts.

It is recognized that, the method adopted for the catalyst preparation is a critical factor determining its physico-chemical properties, dictating the interaction between the active components and strongly afecting its catalytic activity [28,29]. On the other hand, it is well known that, several advantages distinguish the sol-gel procedure including a simple technology, low processing temperature and flexible control of the structure and size of final products via many operating parameters [30,31]. Nevertheless, the evaluation of the NH_3-SCR activity of an aerogel WO_3-CeO_2-TiO_2 derived sol gel system has not yet been studied. The aim of this work was the synthesis and characterization of new mesoporous and well-structured cerium based aerogel catalysts (CeO_2-TiO_2, WO_3-TiO_2 and WO_3-CeO_2-TiO_2) for the NH_3-SCR reaction in a wide temperature range (150-500 °C) under O_2 rich conditions and at relatively high gas hourly space velocity "GHSV" (120.000 h^{-1}).

2. Experimental

2. 1. Preparation of aerogel catalysts

The TiO_2, CeO_2-TiO_2, WO_3-TiO_2 and WO_3-CeO_2-TiO_2 aerogel samples were synthesised via "the one step sol-gel method-supercritical drying approach" according to the same procedure reported in our previous works [32,33]. So, for the preparation of pure titania, a suitable quantity of Ti precursor (Ti(IV) isopropoxide: $Ti(O_iC_3H_7)_4$, Sigma-Aldrich, 97%) was dissolved, at room temperature and under continuous stirring, in an appropriate volume of anhydrous ethanol (C_2H_6O, Aldrich, 99.8 %) used as a solvant. Then, the needed amount of a complexing agent (Ethyl acetoacetate, $C_6H_{10}O_3$, Fluka, > 99.5%), yielding a molar ratio nEtacac/nTi =1, was introduced into the mixture to control the reactions rate. After ageing under stirring, a dilute solution of HNO_3 (0.1 M) was gradually added, with a molar ratio nH_2O/nTi = 10, to accomplish hydrolysis, and the slurry was keep under stiring until the gelification. Finally, the resulting gel was transformed into TiO_2 aerogel powder by drying in an autoclave under the supercritical drying conditions of the ethanol (T = 243 °C and P = 63 bar). All the catalysts were elaborated using the same methodology; cerium (III) nitrate hexahydrate ($Ce(NO_3)_3$,6 H_2 O, Sigma-aldrich, ≥ 98.5 %) and ammonium metatungustate hydrate ($(NH_4)_6H_2W_{12}O_{24}.xH_2O$, Sigma Aldrich, > 99.9%) were used as Ce and W sources, respectively. Their calculated amounts, giving a theoretical loadings of 10 % wt. CeO_2 and 10 % wt. WO_3, were introduced in the slurry before the hydrolysis step (a few quantity of $H_2C_2O_4.2H_2O$ aqueous solution (0.1 M) was employed to facilitate the dissolution of W precursors before use). All the obtained aerogel powder were calcined for 3 h at 500 °C under O_2 flow (30 mL min^{-1}).

2. 2. Characterization of aerogel samples and catalytic test

Powder X-ray diffraction analyses were done on a Brüker AXS D8 diffractometer with a CuKα radiation (λ = 1.5406 Å). The data of 2θ were collected from 2 to 80 ° with a step size of 0.02 °. The crystallites size (D) of powder aerogels was calculated using the Scherrer formula [34] based on the most intense peak of TiO_2 anatase phase (~2θ = 25 °): D = 0.89 (λ /βcosθ), where, λ is the wavelength of XR radiation, β is the corrected peak width at half-maximum intensity (FWHM in radians), and θ is the peak position of the main reflection.

Textural properties of powder aerogels (BET surface area, total pore volume, average pore diameter and pore size distribution) were determined by multipoint N_2-adsorption-desorption method, at liquid N_2 temperature (77 K), using an ASAP 2020 (Micromeritics) instrument. Before measurements, the sample was degassed at 200 °C for 6 h to evacuate any adsorbed moisture. The specific surface area was calculated by the Brunauer-Emmett-Teller (BET) equation, while, the pore volume, average pore diameter and pore size distribution were determined by Barrett- Joyner-Halenda (BJH) method from the desorption branches of the isotherms.

Diffuse reflectance ultraviolet visible spectroscopy (DR-UV-vis) was carried out over the powder samples, at room temperature, in the 200-900 nm range with a speed of 960 nm min^{-1} and an aperture of 4 nm. PerkinElmer spectrophotometer type lambda 45 equipped with an integrating sphere type RSA-PE-20 was used as apparatus.

Temperature Programmed Desorption by Ammonia (NH_3-TPD) was performed using an AUTOCHEM 2920 (Micromeritics) equipped with a TCD detector. Prior to NH_3 adsorption, the catalyst (m= 0.05 g of powder) was activated under air flow (30 mL min^{-1}) at 500 °C (ramp 10 °C min^{-1}) for 30 min. After cooling to 100 °C, the sample was saturated with ammonia (5 vol% NH_3 in He, flow rate = 30 mL min^{-1}) for 45 min. Then, it was flushed with He (30 mL min^{-1}) during 2 h to remove physisorbed NH_3. Finally, the ammonia was desorbed in He flow (30 mL min^{-1}) from 100 °C to 600 °C using a heating rate of 10 °C min^{-1}.

Temperature Programmed Reduction by Hydrogen (H_2-TPR) was realized on a Micromeritics AUTOCHEM 2910 equipped with a TCD detector. Briefly, the aerogel (m = 0.05 g of powder) was pre-treated under 5 vol % O_2 in He (flow rate = 30 mL min^{-1}) at 500 °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 mL min^{-1}) then exposed to a flow containing 5 vol % H_2 in Ar (30 mL min^{-1}) and heated between 50 and 800 °C (10 °C min^{-1}).

The selective Catalytic Reduction (SCR) tests were carried out over the catalysts in a fixed-bed quartz flow reactor operating at atmospheric pressure. The reaction conditions were controlled as follows: 400 ppm NO, 400 ppm NH_3, 8 % O_2 in He as a balance gas and 100 mL min^{-1} total flow rate yielding a gas hourly space velocity (GHSV) of 120,000 h^{-1}. Before measurements, the catalyst (0.05 g of powder) was activated in situ at 200 °C for

30 min under O_2/He (20/80, v/v) flow then cooled to 100 °C. Afterward, the NO conversion curves were recorded between 100 and 500 °C with the heating rate of 6 °C min^{-1}. The reactants and products gases were continuously analyzed by an online Pfeiffer Omnistar quadruple mass gas spectrometer equipped with Channeltron and Faraday detectors (0-200 amu).

3. Results and discussion

3. 1. Structural properties of aerogel catalysts

The results of X-ray diffraction analysis are exposed in Fig. 1 and table 1. As shown in Fig. 1, all the samples are well structured materials and develop essentially the diffraction peaks of TiO_2 anatase phase at 2θ ≈ 25.3° (hkl:101); 36.9°(103); 37.8° (004); 38.6° (112); 48.2° (200); 53.9° (105); 55.2° (211); 62.7° (204); 69.0° (116); 70.4° (220) and 75.2° (215) [ICSD 01-083-2243]. Additional peaks with a low intensity are observed at 2θ ≈ 28.5° (hkl: 111); 32.9° (200) in the diffractogram of samples containing cerium and are attributed to a low amount of the CeO_2 cubic phase [ICSD 00-034-0394]. However, no reflections related to WO_3 crystalline phase (most intense one at 2θ ≈ 23,7° (hkl: 110) [ICSD 01-089-8053]) are detected in the case of solids containing W revealing the high dispersion state of tungsten species at the catalysts surface. This result agreed with that obtained by Li et al. [35] for CeO_2-WO_3/TiO_2 SCR catalyst prepared by the impregnation method, containing 10% WO_3 and 4 wt. % CeO_2. The authors concluded that the WO_3 cristallites are in the form of "monolayer" species, constituted by W_xO_Y surface complexes, that cover the TiO_2 surface more or less completely, and are undetectable via XRD.

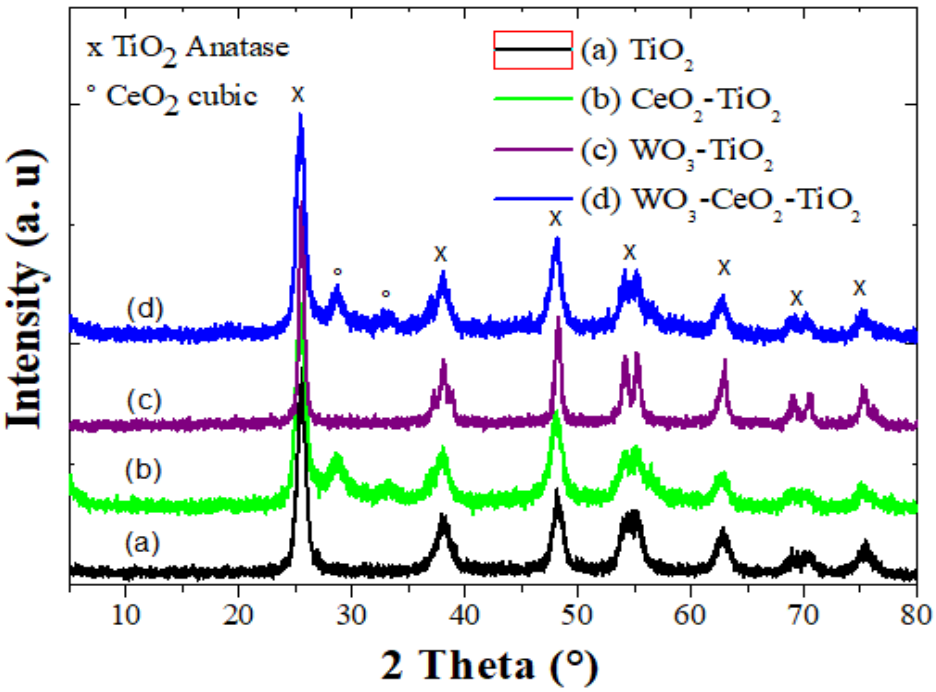

Figure 1. Powder XRD patterns of nanostructured mesoporous aerogel catalysts.

The crystallites size of each sample is given in table 1 which demonstrates that all the catalysts are classified as nanostructured materials with a nanometer size in the range of 8-15 nm. It should be mentioned that, there are no obvious changes in the crystallites size of titania for CeO_2-TiO_2 and WO_3-CeO_2-TiO_2 solids. However, the addition of tungsten species only contributes to the growth of the TiO_2 cristallites size (from 8.6 for TiO_2 to 15 nm for WO_3-TiO_2). According to the literature reports, this phenomen could be explained by (i) the agglomeration of the metal clusters to form larger particle size [36] or (ii) a sintering which probably happened in the WO_3-TiO_2 sample and has led to the increase of the TiO_2 grain size [37].

Table 1. XRD phases and TiO_2 crystallites size of nanostructured mesoporous aerogel catalysts.

Sample	XRD phases	FWHM (°)	TiO_2 crystallites size D (nm)
TiO_2	Anatase	0.892	8.6
CeO_2-TiO_2	Anatase + ceria	0.884	8.6
WO_3-TiO_2	Anatase	0.511	15.0
WO_3-CeO_2-TiO_2	Anatase + ceria	0.931	8.2

3. 2. Textural properties of aerogel catalysts

The porous structure of catalyst is very important for catalytic activity. Fig. 2 shows the N_2 adsorption-desorption isotherms at 77 K and pores size distribution curves of the different solids. As it can be seen, all the samples exhibit a type IV isotherm, characteristic of mesoporous materials according to the IUPAC classification [38], and are characterized by a unimodal porous distribution with different hysteresis loops. Hence, TiO_2 and CeO_2-TiO_2 solids develop a H2 type of hysteresis loops demonstrating the presence of ink-bottle pores in their mesoporous texture [39], while, the presence of cylindrical mesoporous channels in the case of WO_3-TiO_2 and WO_3-CeO_2-TiO_2 catalytic systems is verified based on their hysteresis loops which were identified as H1 type [40].

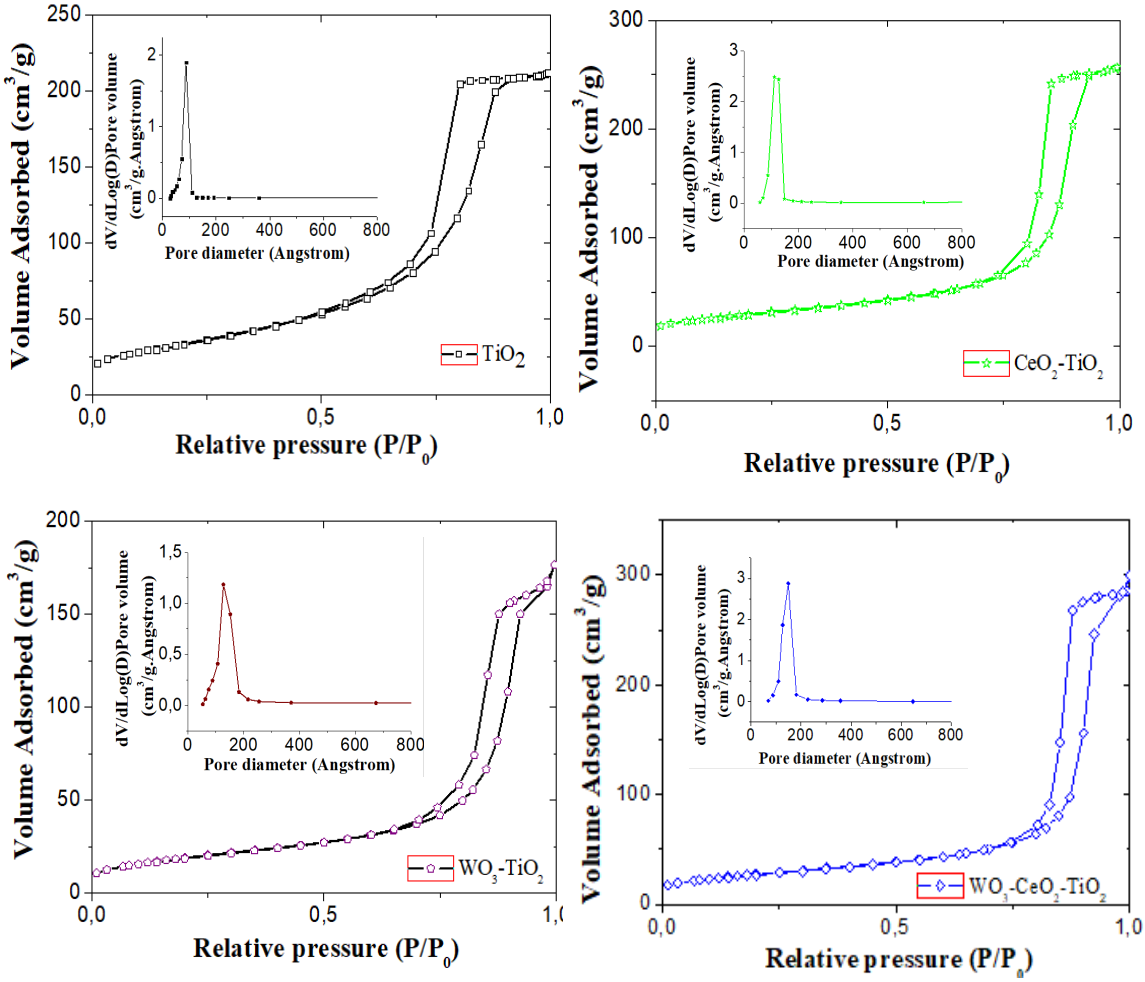

Figure 2. N_2 Adsorption-desorption isotherms and pore size distribution curves of nanostructured mesoporous aerogel catalysts.

It is established that large specific surface area implies more active sites exposed on the surface of catalyst and more contact opportunities with gas molecules, which can promote the SCR activity [41]. As revealed in table 2, all the aerogel catalysts, calcined at 500 °C, are characterized by a developed mesoporous texture with a medium average pore diameter < 145 Å, high surface area ($70 \leq S_{BET} \leq 122$ m^2/g) and large porosity ($0.27 \leq V_{PT} \leq 0.46$). This will be beneficial for the catalytic behaviour. It is important to note that the decrease of the surface area of TiO_2 after Ce and/or W incorporation can be explained by the blockage of some pores of titania carrier by the supported active species [32,33]. According to Petrović et al. [37], the growth of the TiO_2 crystallites size after W addition contributes also to the decrease of the specific surface area and total pore volume of WO_3-TiO_2 solid which is characterized by the lowest values of textural parameters (S_{BET} and V_{PT}, table 2) but by the highest one of the TiO_2 crystallites size (table 1).

Table 2. Textural properties of nanostructured mesoporous aerogel catalysts.

Sample	BET surface area (m^2/g)	Total pore volume (cm^3/g)	Average Pore diameter (Φ_{pore} , Å)
TiO_2	122	0.33	79
CeO_2-TiO_2	106	0.40	117
WO_3-TiO_2	70	0.27	133
WO_3-CeO_2-TiO_2	99	0.46	145

3.3. Characterization of supported active species

The valence states and environment coordination of Ce and W species at TiO_2 surface are examined via Diffuse-Reflectance UV-vis spectroscopy and the results are illustrated in Fig. 3. The TiO_2 support shows a typical absorption band in the 200-400 nm range with a maximum at around 217, 275 and (320 ; 350) nm, corresponding to $O^{2-} \rightarrow Ti^{4+}$ charge transfert transitions in tetrahedrally coordinated Ti^{4+}, octahedrally coordinated Ti^{4+} and TiO_2 anatase phase, respectively [32,33,42]. Additional band appear at $\lambda > 350$ nm in the UV vis spectra of CeO_2-TiO_2 and WO_3- CeO_2-TiO_2 catalysts and are ascribed to UV absorptions of cerium species. In fact, it was reported that cerium species are characterized by $O^{2-} \rightarrow Ce^{3+}$ and $O^{2-} \rightarrow Ce^{4+}$ charge transfer bands (CT) in the 220-250 and 280-400 nm ranges, respectively [32]. The typical absorption bands of tungsten species are usually observed in the 210-270 nm range for the isolated tetrahedral W^{6+} and oligomerized W species [43-46] and between 300 and 375 nm for the polymeric octahedral W^{6+} species [40, 43-46]. For the WO_3-TiO_2 and WO_3-CeO_2-TiO_2 solids, the UV absorption bands of tungsten species seem to be overlapped by those associated to Ti and Ce species. The absence of UV characteristic absorptions of crystalline WO_3 (normally in the range of 430-450 nm [46]) confirms, in line with the XRD results and in perfect agreement with our previous results [47], the highly dispersion state of tungsten species at the catalysts surface.

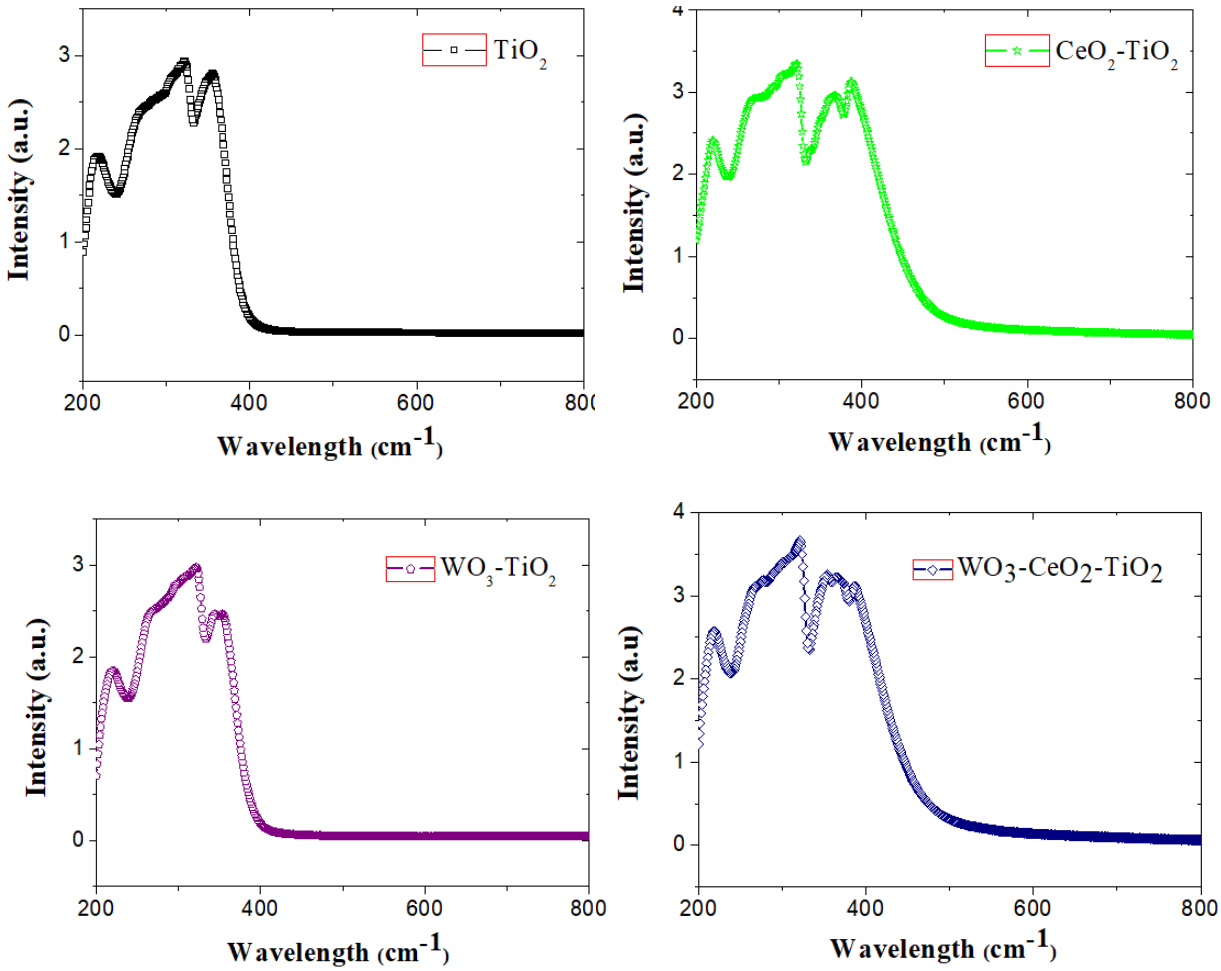

Figure 3. DRUV-Vis spectra of nanostructured mesoporous aerogel catalysts.

3.4. Acidic performance of aerogel catalysts

The acidic properties of catalysts are studied by means of the NH_3-TPD technique. The results are depicted in Fig. 4. As exposed in the NH_3-TPD curves of samples, all the aerogel materials display a broad peak from 100 to 400 °C which could be attributed to NH_3 desorbed from weak and medium acid sites [32,33]. The intensity of this peak decreases following this order: $TiO_2 > CeO_2\text{-}TiO_2 > WO_3\text{-}TiO_2 > WO_3\text{-}CeO_2\text{-}TiO_2$. This could indicate the existence of diverse interactions between the support and the supported active species which cover some acidic sites at the surface of solids. Similar results, demonstrating the decrease of the surface acidity of Mn-based catalysts after cerium addition, has been obtained in our previous work and was correlated with the existence of Mn-Ce and Ce-SO_4^{2-} interactions which inhibit some acidic sites of solids [48]. Remarkably, a high temperature NH_3 desorption peak (at T = 440 °C) is observed in the NH_3-TPD profile of $WO_3\text{-}TiO_2$ only and could be ascribed to the ammonia desorbed from new strong acid sites created by tungsten species at the titania surface. In fact, Sohn et al. [49] mentioned that the combination of TiO_2 and WO_3 can generate stronger acid sites and more acidity as compared with the separate components.

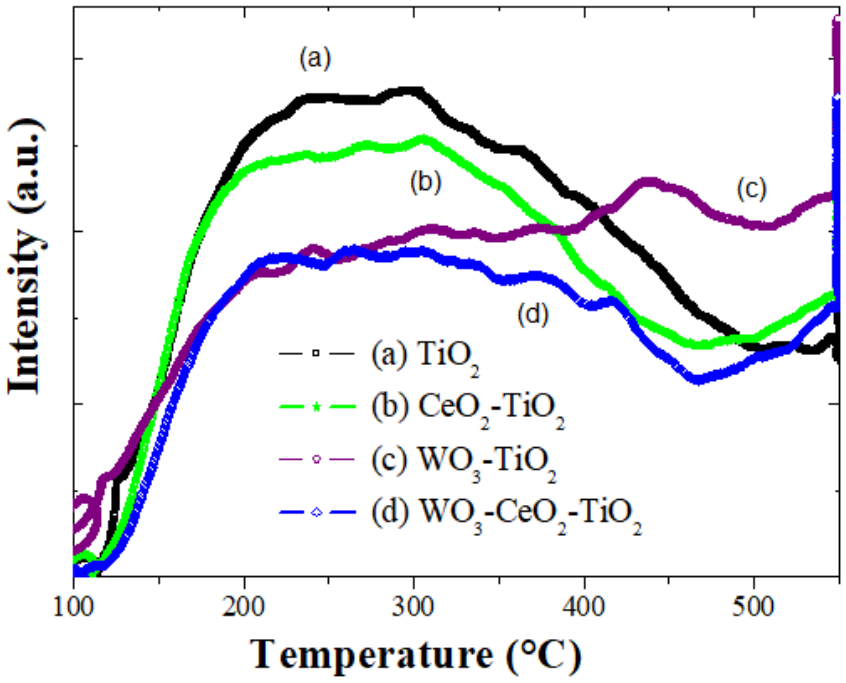

Figure 4. NH_3-TPD profiles of nanostructured mesoporous aerogel catalysts.

3.5. Redox performance of aerogel catalysts

H_2-TPR is a widely used technique in the study of redox properties of catalysts. The H_2-TPR profiles of prepared solids are displayed in Fig. 5.

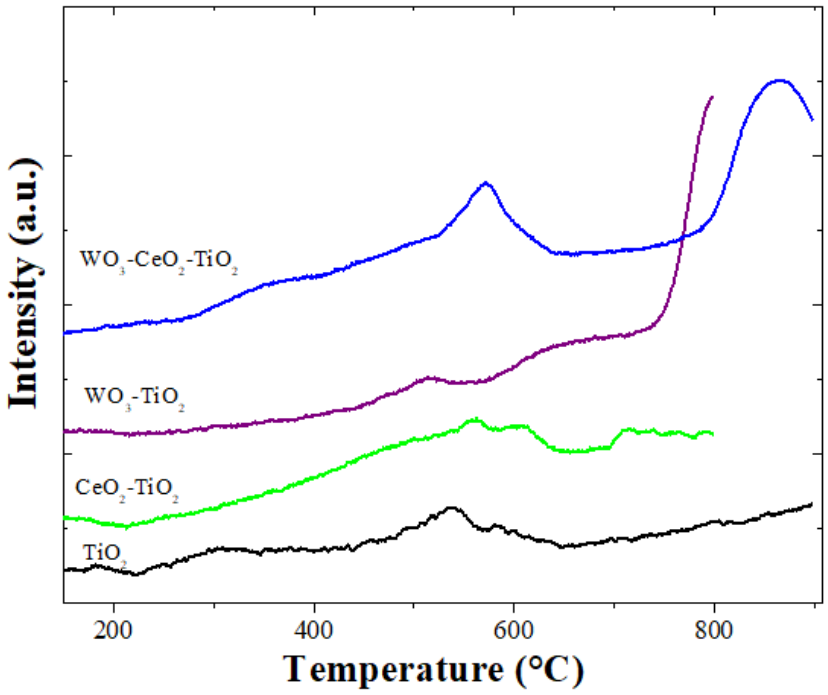

Figure 5. H_2-TPR profiles of nanostructured mesoporous aerogel catalysts.

As illustrated in Fig. 5, the H_2-TPR profile of CeO_2-TiO_2 catalyst shows hydrogen consumption peaks between 400 and 600 °C which are attributed, according to the literature reports, to the reduction of the surface-capping oxygen of stoichiometric ceria of Ce^{4+}-O-Ce^{4+} and cerium species (from Ce^{4+} to Ce^{3+}) [4,32,47,50-52]. The H_2 consumption peaks detected at T > 600 °C for WO_3-TiO_2 sample are related to the reduction of tungsten species [47, 53-55]. For the ternary WO_3-CeO_2-TiO_2 catalyst, the broad peak located in the 300-500 °C temperature range is ascribed to the reduction of surface oxygen of CeO_2 [4,47,50,52], whereas, the peak centered at 570 °C correspond to the reduction of cerium species from Ce^{4+} to Ce^{3+} [32,47,53]. Noticeably, these former peaks are more large and intensive than these obtained in the case of the binary CeO_2-TiO_2 indicating the presence of more surface oxygen and Ce redox sites at the surface of ternary WO_3-CeO_2-TiO_2 aerogel catalyst. Based on the above result, it can be suggested the existence of Ce-W interactions which probably improves the dispersion of cerium species leading to the creation of more surface oxygen and Ce redox sites exposed at the surface of ternary catalyst. It is also worth noting that, the reduction of tungsten species occurs at higher temperature in the presence of Ce (800 °C for WO_3-TiO_2 and 860 °C for WO_3-CeO_2-TiO_2). This confirms the existence of strong interactions between cerium and tungsten species.

3. 6. Evaluation of the SCR activity of aerogel catalysts

Fig. 6 shows the NO conversion as function of the reaction temperature over all the aerogel catalysts. As shown, the TiO_2 support is inactive in the low temperature NO-SCR by NH_3 (T< 300 °C) and displays a maximum of ~ 50 % NO conversion into N_2 at 500 °C. The addition of tungsten slightly increases the NO conversion into N_2 which exceeds 50 % at high temperature (between 415 and 500 °C). The enhancement of the high temperature SCR activity of WO_3-TiO_2 solid can be essentially related to the reactivity of strong acid sites generated by the presence of tungsten species at TiO_2 surface as revealed by the NH_3-TPD analysis. The CeO_2-TiO_2 solid exhibits much higher NO-SCR activity than that of TiO_2 and WO_3-TiO_2, particularly at low temperature (in the 200 and 400 °C temperature range). In fact, the NO conversion obtained over this material increases with the increase of the reaction temperature as follow: 30 % at 250 °C, 76 % at 300 °C and > 90 % between 330 and 400 °C. This high catalytic activity can be mainly correlated with the presence of actives surface oxygen and redox sites generated by the presence of cerium species at the CeO_2-TiO_2 surface, as demonstrated by the H_2-TPR technique. The SCR activity is slightly increased in the case of WO_3-CeO_2-TiO_2 ternary system if compared to CeO_2-TiO_2 binary. Therefore, the NO conversion into N_2 obtained over this new ternary catalyst is 50 % at 250 °C, 85 % at 300 °C and > 90 % between 320 and 400 °C. Taking into account the results of the H_2-TPR analysis, it can be suggested that the strong interactions Ce-W play a key role in enhancing the surface oxygen concentration and

redox properties of WO_3-CeO_2-TiO_2 system and, consequently, increases its reactivity in the NO-SCR reaction. Noting that the decrease of the NO conversion at high temperature over CeO_2-TiO_2 and WO_3-CeO_2-TiO_2 catalysts can be explained by the absence of high temperatures active acidic sites at their surface. In fact, it is established that both acidic and redox properties of catalyst are the key factors which govern the NO-SCR by NH_3 reaction [56]. It is believed that the surface acidity play an important role in the adsorption and activation of ammonia, especially at high temperature but not at low temperature where the reactivity of catalyst is controlled by the redox properties [56].

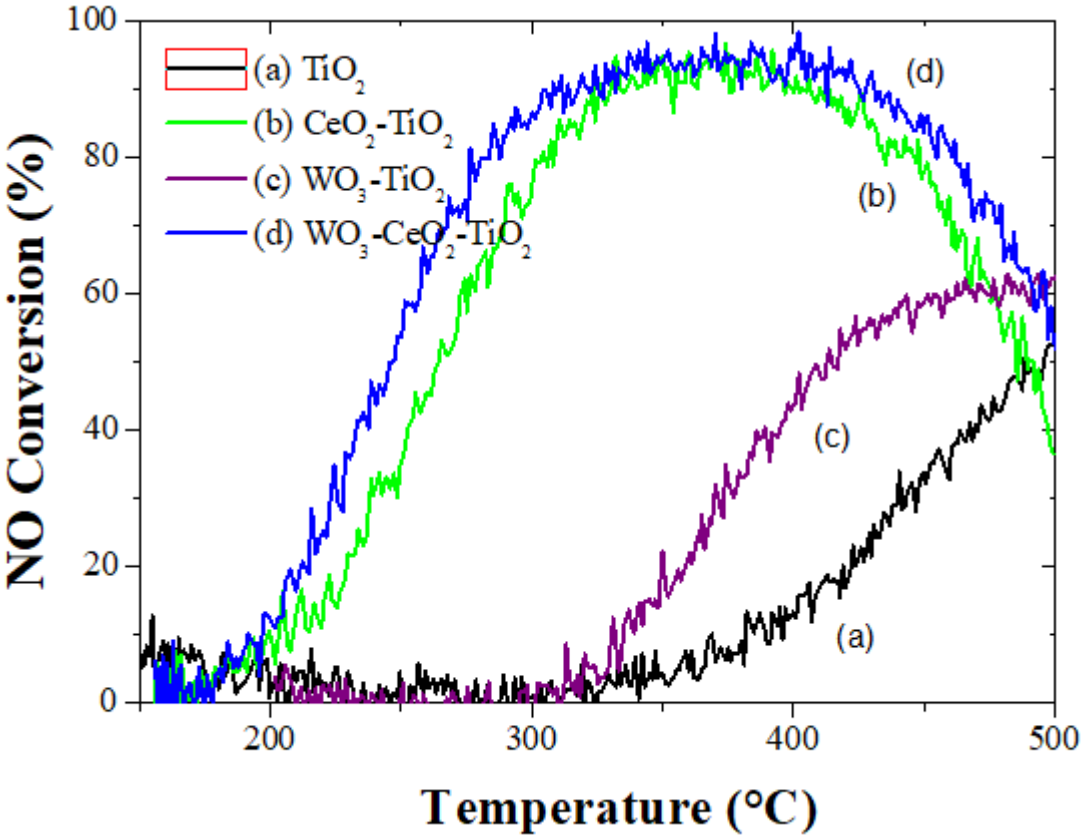

Figure 6. NO removal efficiencies over the nanostructured mesoporous aerogel catalysts: [NO] = [NH_3] = 0.04 %; [O_2] = 8.00 % and balance with He, (GHSV) = 120 000 h^{-1}.

4. Conclusion

New well-structured and mesoporous aerogel catalysts (CeO_2-TiO_2, WO_3-TiO_2 and WO_3-CeO_2-TiO_2) with a nanometer size were elaborated in this study using the sol gel method combined to supercritical drying approach. The solids were characterized and tested in the selective catalytic reduction of NO by NH_3. The developed mesoporous textural and highly crystalline structure of all the samples were proved through the N_2-adsorption-desorption and XRD techniques. The NH_3-TPD and H_2-TPR analyses demonstrated that adding cerium or tungsten species contributes to the decrease of the total acidity of titania carrier but leads to the creation of

diverse redox sites at its surface. CeO_2-TiO_2 catalyst was found more active in the NH_3-SCR of NO than WO_3-TiO_2 one, especially at low temperatures, due to the presence of reactive surface oxygen and active Ce redox sites at its surface. The simultaneous presence of Ce and W and the strong interactions developed between them affect the surface properties of WO_3-CeO_2-TiO_2 catalyst, especially its redox behavior, and increases its NO-SCR activity. This new aerogel catalyst help to reduce more than 90 % NO into N_2 between 320 and 400 °C.

Declarations

Funding (Not applicable)

Conflicts of interest : There are no conflicts of interest to declare

Availability of data and materials (Not applicable)

Code availability (Not applicable)

Authors contributions (Not applicable)

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