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Preparation of extruded catalysts based on TS-1 zeolite for their application in propylene epoxidation

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ABSTRACT

In this work, the extrusion process of titanium silicalite (TS-1) zeolite has been investigated and optimized in order to obtain materials with convenient catalytic and mechanical properties for their application in the propylene epoxidation with hydrogen peroxide using fixed bed reactors. Thereby, a variety of TS-1 samples were prepared having particle sizes comprised between 0.2 and 0.7 μm. It has been observed that the increase of the zeolite particle size affects negatively the mechanical strength of the extrudates. Likewise, a variety of inorganic binders such as clays or inorganic silica have been explored; the best results in terms of minimizing the H_2O_2 decomposition being obtained with sepiolite. Moreover, three extrusion methods and different TS-1/inorganic binder mass ratios were investigated in order to achieve an extruded material with satisfactory mechanical strength. The optimum extruded catalyst was prepared using a TS-1/sepiolite mass ratio of 60%/40% and employing ultrasounds during the paste preparation. Finally, the catalytic activity of the optimum extruded TS-1 zeolite has been evaluated for the propylene epoxidation reaction using a fixed bed reactor, achieving conversion and H_2O_2 selectivity values close to 97% and 80%, respectively.

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1. Introduction

Propylene oxide (PO) is one of the most important starting materials in the chemical industry. It is widely used for the manufacture of polyurethane foams, coatings, seals and adhesives in the automotive and housing industries. In the last years, the PO market volume has undergone a substantial growth, increasing from 5.1 million metric tons in 2003 to 6.3 million metric tons in 2007. Currently, there are two main industrial methods to obtain propylene oxide. The chlorohydrin process is based on a two-step reaction. Firstly, propylene is reacted with chlorine and water leading to the formation of chlorohydrin as an intermediate. The chlorohydrin solution is subsequently dehydrochlorinated with calcium hydroxide in a distillation tower giving PO as product overhead the column. This process, however, presents important environmental and safety drawbacks since chlorine is a toxic and corrosive reactant and large quantities of harmful by-products and wastes (40 tons of $CaCl_2$ containing wastewater per ton of PO) are generated. In addition, production of PO by this procedure involves multiple steps and it is energy-intensive consuming. The second industrial method for PO production is the so-called hydroperoxide process. In this case, ethylbenzene or isobutane are reacted in a first step with oxygen to produce an organic hydroperoxide. This latter is subsequently used to oxidize propylene giving PO, which is then purified. The major inconvenience of the hydroperoxide process is that large amounts of co-products are generated, such as styrene or tert-butyl alcohol, depending on the initial reactants employed. The formation of co-products may represent up to three times the PO production. Therefore, the viability of this process depends on the existence of a market for such co-products [1].

New PO technologies without by-products generation are now coming close to commercialization [2]. For this purpose, most efforts have been focussed on the direct conversion of propylene to PO using a suitable catalyst and a source of oxygen. In particular, the epoxidation of propylene with diluted H_2O_2 over titanium silicalite (TS-1) has attracted much attention. Using TS-1 as epoxidation catalyst of propylene leads to production of water as the unique by-product, along with a high activity and selectivity, achieving a nearly total H_2O_2 conversion in a methanol-based solvent system. These advantages have made propylene epoxidation over TS-1 zeolite, in the last years, as the best alternative to

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manufacture PO. However, the industrial implementation of this process has been hindered by the relatively high cost of H_2O_2. This problem can be solved by the production of H_2O_2 in the own manufacturing plant, mainly using the anthrahydroquinone autooxidation process [3].

Concerning the TS-1 catalyst, this is a well-known titanium-substituted zeolite belonging to the family of materials with MFI structure. After the first report on the TS-1 synthesis by Taramasso et al. [4], this zeolite has been found to be an excellent catalyst for a large variety of oxidation reactions under mild conditions using H_2O_2 as oxidant [5–8]. The exceptional catalytic activity of TS-1 is mainly due to the following three features of the Ti sites: hydrophobic environment, tetrahedral coordination of the Ti centres and constrained reaction sites. For these reasons, TS-1 zeolite has become one of the most relevant oxidation catalysts in the last 20 years [9].

For their industrial application in a fixed bed reactor, TS-1 zeolite powders must be shaped into bodies such as granules, spheres and extrudates, in order to decrease the pressure drop and to achieve a convenient mechanical strength. However, the difficulty to mold the powdery TS-1 represents an obstacle for its use in fixed bed reactors [10]. In general, an extruded material is manufactured from a paste, which is previously obtained by blending the catalyst powder with the following components: (a) an inorganic binder, which provides the mechanical strength, (b) an organic binder, which increases the viscosity of the paste in order to get a suitable plasticity during extrusion and (c) a liquid phase to provide effective lubrication during the extrusion [11].

In spite of the huge amount of literature that has been published about the synthesis, properties and catalytic applications of TS-1 zeolite [12–16], almost no information can be found in the open literature about its agglomeration. However, this is an essential step regarding its commercial application. In this context, the aim of the present work has been the development of a process for the extrusion of TS-1 zeolite, so the resulting material can be applied as a catalyst in propylene epoxidation using a fixed bed reactor. In order to get the most suitable catalyst, TS-1 samples prepared from different synthesis methods, several inorganic binders and three extrusion methods have been explored. The catalytic activity of the extruded TS-1 sample has been evaluated for the propylene epoxidation reaction using a fixed bed reactor.

2. Experimental

2.1. Preparation of TS-1 samples

Five TS-1 zeolite samples have been prepared using different synthesis methods described previously in literature:

(1) TS-1(1) and TS-1(2) samples were prepared according to the conventional method developed by Taramasso et al., using tetraethylorthosilicate (TEOS, from Aldrich) and tetraethylorthotitanate (TEOT, from Aldrich) as silica and titanium sources, respectively [4]. The former sample was synthesized using conventional heating at 170 °C for 24 h and the latter one was obtained by applying microwave radiation at 170 °C for 2 h.

(2) TS-1(3), TS-1(4) and TS-1(5) samples were synthesized by wetness impregnation of an amorphous SiO_2–TiO_2 xerogel [12]. Samples TS-1(4) and TS-1(5) were crystallized under microwave radiation using two different heating programs. The former was prepared using a single heating at 170 °C for 4 h. The latter was obtained by a first heating at 100 °C for 18 h followed by a second heating at 170 °C for 3.5 h [14]. TS-1(3) sample was synthesized using conventional heating at 170 °C for 24 h in a static reactor.

2.2. Preparation of extruded catalysts based on TS-1 zeolite

The inorganic binders considered in this work were as follows: bentonite (Sud Chemie), montmorillonite (Across), sepiolite Pansil (Tolsa), sepiolite Pangel (Tolsa), silica PQ (Grace Davison), silica gel (J.T. Baker) and silica spheres (Saint Gobain Norpro).

The extrusion process employed in this work is based on the generation of a homogeneous paste comprised by the following constituents: (a) the inorganic binder, which provides the suitable physical strength, (b) methylcellulose polymer as organic binder, providing the appropriate plasticity to the paste, (c) TS-1 zeolite powder, as the catalyst and (d) distilled water.

Three different extrusion procedures were employed consisting in the introduction of modifications in the paste preparation step. In method 1, the paste was obtained by dry-mixing of the clay (sepiolite), methylcellulose and TS-1 powder. Then, distilled water was added to the powder mixture (200 wt% of water relative to the solid content) giving the final paste. Method 2 consisted in the clay addition to a suspension of TS-1 powder in water (200 wt% of solid), methylcellulose being subsequently added. Method 3 was analogous to method 2, but using ultrasounds and larger water amounts (240 wt% of solid) to generate the TS-1 powder suspension. The use of ultrasound is expected to disperse possible TS-1 powder aggregates, thus facilitating the formation of a more homogeneous paste when the clay was added (particles smaller than 5 μm tend to form aggregates) [15].

Different pastes were obtained using TS-1 powder/sepiolite mass ratios from 90/10 to 50/50 while keeping the amount of methylcellulose as 10 wt% of the total solid weight. Once the paste was obtained and well mixed, it was loaded into a vertical ram extruder, shown in Fig. 1. Then, a piston was placed into the extruder and it was pushed straightforward until the paste passed through a circular die (3 mm diameter). The shaped-rod products were cut in 2 mm pieces and dried in a chamber under controlled temperature and humidity conditions (80 °C and 10% of humidity). Thereafter, the solids were calcined at 650 °C under static air, using a slow temperature ramp in order to gradually remove the water, to burn off the organic content and to enhance the mechanical strength of the extrudates.

2.3. Characterisation

All the samples obtained were characterised by a number of techniques. X-ray diffraction (XRD) patterns were collected with a Philips X'PERT MPD diffractometer using Cu Kα radiation with step size and time of 0.02° and 10 s, respectively. The coordination of the titanium atoms, as well as the possible presence of extra-framework TiO_2 phases in the samples were determined by diffuse reflectance UV-Vis spectroscopy (DR UV-Vis) under room conditions using a CARY-500 spectrophotometer equipped with a diffuse reflectance accessory. Titanium contents of the synthesized samples were determined by means of atomic emission spectroscopy with induced coupled plasma (ICP-AES) analyses, which were performed in a Varian Vista AX instrument. Previously to the analysis, samples were dissolved by an acid-digestion treatment. SEM images were obtained in a XL30 ESEM FEI (Philips) electron microscope. TEM images were recorded in a Philips Technai 20 electron microscope operating at 200 kV.

The resistance to crushing of the extruded materials was measured by the standardized Shell Test (SMS 1471). However, due to the relatively large amount of sample required for this analysis ($\approx$20 cm^3 of catalyst), the mechanical strength of the extrudates over individual particles was also evaluated through the application of different weights until these particles were destroyed. Thus, it is possible to calculate the maximum pressure

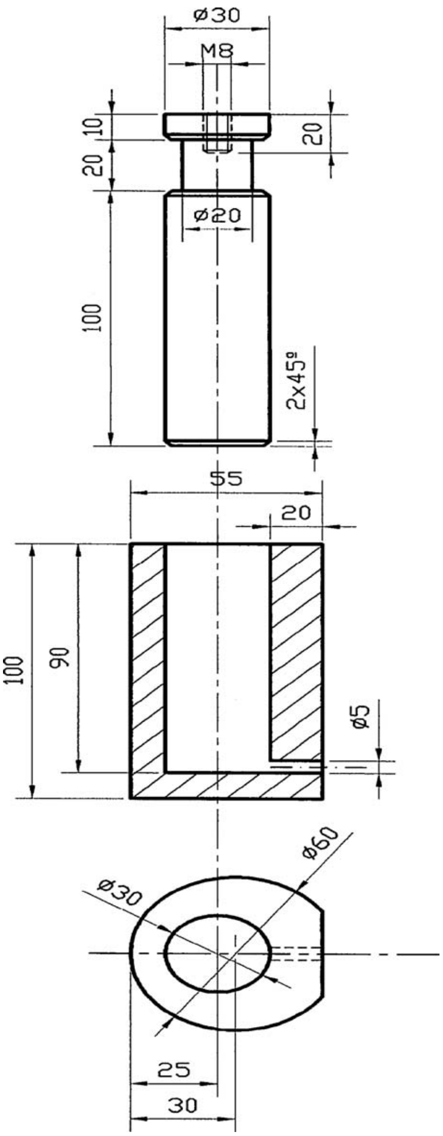

Fig. 1. Extruder ram scheme.

that the particle sample can support. This measurement of the mechanical strength over one particle was carried out by applying the weight using two different particle configurations, as shown in Fig. 2.

2.4. Catalytic tests

The catalytic activity of the TS-1 powder samples was evaluated through the 1-octene epoxidation reaction using H_2O_2 as oxidizing agent. This reaction was performed into a round-bottomed flask fitted with a condenser, a thermometer and a magnetic stirrer. Methanol was used as solvent. Typically, the weight proportions employed were 26 wt% of 1-octene (Aldrich), 72 wt% of methanol (Baker) and 0.9 wt% of H_2O_2. The reaction mixture was heated up to 333 K and then, 1 g of pure zeolite was added. The duration of the reaction experiment was 1.5 h. The same procedure was employed to check the non-efficient decomposition of hydrogen peroxide produced by the inorganic binders.

The catalytic activity of the optimum extruded sample was evaluated using the propylene epoxidation reaction in a fixed bed reactor. The length and the inner diameter of the fixed bed reactor were 1 m and 0.0175 m, respectively. The amount of the extruded catalyst loaded to the reactor was 32 g (TS-1 powder/sepiolite mass ratio: 60%/40%). The catalyst was mixed with carbon silicide in order to ensure a homogeneous flow of the reactants. Typical reaction conditions were as follows: temperature, 323 K; pressure, 3 MPa; solvent, methanol; initial H_2O_2 weight concentration, 9%; propylene:H_2O_2 molar ratio, 2:1; space velocity WHSV, 0.33 h^{-1}. Propylene and H_2O_2 diluted in methanol were separately fed into the reactor.

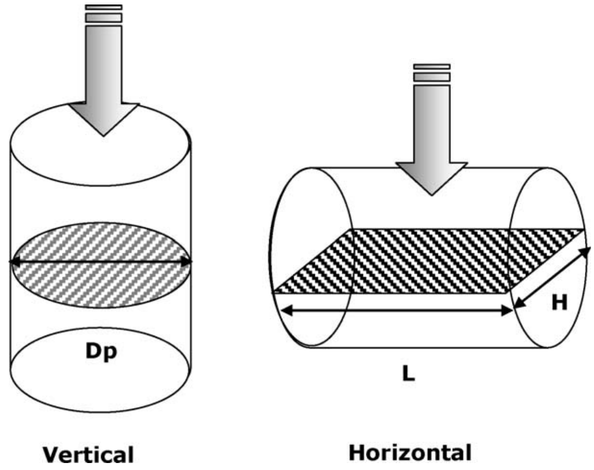

Fig. 2. Mechanical strength evaluation over an individual particle.

Reaction products were analyzed by gas chromatography (HP6850 GC, capillary column HP-INNOWax 60 m × 0.25 mm × 250 mm, FID detector). H_2O_2 conversions were calculated by standard iodometric titration. Propylene glycol (PG) and mono-methyl ethers were detected as the main by-products.

3. Results and discussion

3.1. Properties of the powdered TS-1 samples

Five different TS-1 zeolite samples were prepared by using either the conventional method based on the crystallization of a liquid and transparent gel [4] (TS-1(1) and TS-1(2)) or the method developed by our group consisting in the crystallization of SiO_2–TiO_2 xerogels, previously wetness impregnated with a TPAOH solution (TS-1(3) to TS-1(5)) [11]. On the other hand, both conventional heating (TS-1(1) and TS-1(3)) and microwave radiation (TS-1(2), TS-1(4) and TS-1(5)) were applied during the crystallization step. Fig. 3(a) and (b) illustrates the XRD patterns and DR UV-Vis spectra obtained for all the TS-1 zeolite samples. The XRD patterns indicate that all the samples were highly crystalline, showing the characteristic peaks of MFI structure. The DR UV-Vis spectra of the samples show a strong absorption band at 200–220 nm, which corresponds to tetrahedrally coordinated Ti(IV). This result is a confirmation that most of the Ti atoms were isolated into framework positions of the zeolite lattice by all the synthesis methods applied. However, for the TS-1(1) sample, an additional absorption band around 330 nm was detected, indicating that extra-framework TiO_2 phases were also present in this sample.

Table 1 summarizes the Ti content (determined by ICP-AES), the synthesis solid yield and the particle size (determined by SEM) for the different powdered zeolites as a function of the synthesis conditions. It can be observed that all the catalysts obtained present significant Ti amounts, in the range of 2.23–3.15 wt%, expressed as TiO_2. Moreover, except for the sample TS-1(5), the

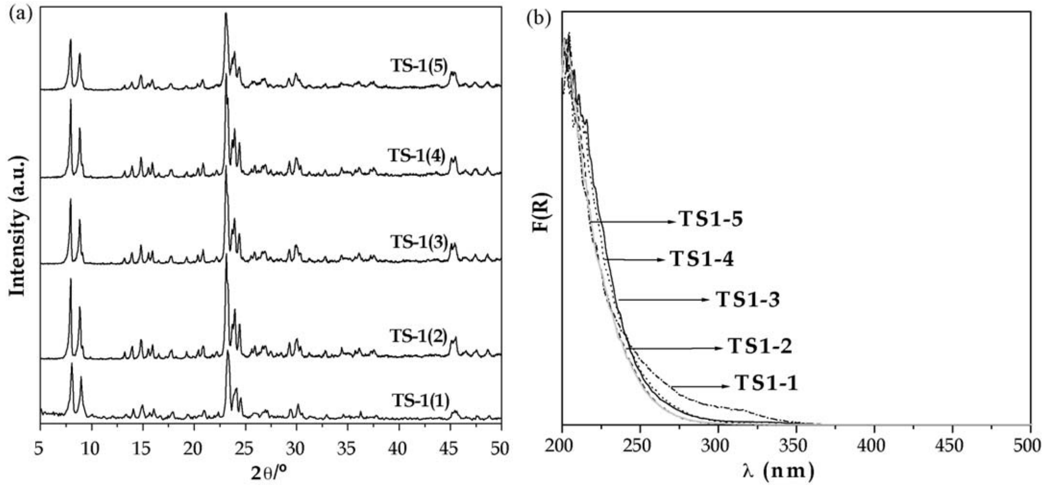

Fig. 3. (a) XRD patterns and (b) DR UV-Vis spectra of calcined TS-1 samples prepared by different methods.

synthesis based on the wetness impregnation of amorphous SiO_2–TiO_2 xerogels led to higher synthesis yields (>90%).

The particle sizes were clearly different depending on the synthesis method employed, showing sizes between 140 and 790 nm. The procedure based on the crystallization of liquid gels led to smaller particle sizes than the wetness impregnation route of SiO_2–TiO_2 xerogels. However, in this case the use of microwave heating radiation allows the particle size to be controlled through the design of the suitable heating program. The particle size of the TS-1 powder sample is an important issue as smaller particle sizes may involve less diffusional restrictions and a higher homogeneity during the blending of the zeolite powder with the binders.

The catalytic activity of the powdered TS-1 samples was evaluated in the 1-octene epoxidation using H_2O_2 as oxidizing agent. Conversion and selectivity results so obtained are summarized also in Table 1. H_2O_2 selectivity has been defined as moles of epoxide formed in regard to the oxidant consumed. The difference to 100% takes into account the H_2O_2 decomposition and the formation to oxygenated by-products. All the catalysts prepared exhibited good activity in 1-octene epoxidation, showing very high H_2O_2 conversions (over 96%) and selectivities around 60%, except TS-1(4) that exhibits a lower selectivity value (50.3%), which is probably due to the presence of small proportion of extra-framework TiO_2 in this sample. Comparing the results obtained with samples TS-1(1) and TS-1 (3), it can be concluded that, the catalytic activity of the samples seems not to be affected by the particle size, at least under the reaction conditions here employed.

3.2. Selection of the inorganic binder

The role of the inorganic binders is to provide the extrudate with satisfactory physical strength, both during its manufacturing process and in the finished product. However, this component may interfere with the catalytic properties of the final extruded particles since additional species and phases are introduced to the composition of the final catalytic system. Consequently, special attention must be paid to the selection of the inorganic binder in order to preserve the catalytic activity of the zeolite. In this way, a study of non-efficient decomposition of hydrogen peroxide was carried out using different materials usually employed as inorganic binders of catalysts. A variety of clays (bentonite, montmorillonite and sepiolite Pangel), as well as different inorganic silica, were evaluated in terms of H_2O_2 decomposition. The resultant values obtained for the different inorganic binders are shown in Fig. 4. All the inorganic binders tested led to very high H_2O_2 decomposition ratios (in the range of 80–99%), except for sepiolite Pangel, which decomposed 55% of H_2O_2. Therefore, this last binder was selected as the most suitable for the following TS-1 extrusion studies. It must be taken into account that, although the H_2O_2 decomposition produced by sepiolite is not negligible in the activity of the final catalyst, when TS-1 zeolite is present in the reaction media, the epoxidation rate is several times higher than the H_2O_2 decomposition rate.

3.3. Effect of the extrusion method

The procedure to manufacture an extruded material involves the following steps: components mixing, production of a paste, controlled drying and thermal processing. The extrusion paste, which is a mixture of solid and liquid phases, must retain its shape for time enough to allow any subsequent processing to be carried out [15]. In this work, the paste was comprised by the inorganic binder (sepiolite), an organic binder (methylcellulose polymer), TS-1 zeolite powder and distilled water. Three different methods for the paste preparation were investigated in order to select that leading to the most homogeneous paste. Method 1 consisted in the

Table 1
Synthesis parameters and properties of TS-1 samples.

Sample	Synthesis method	Heating	TiO_2 (wt%)	Yield (%)	Particle size (nm)	Conversion[a] (%)	Selectivity[b] (%)
TS-1(1)	LG	Conventional	2.55	90	140	98.5	68
TS-1(2)	LG	Microwave	2.23	52.8	430	97.1	68.4
TS-1(3)	XG	Conventional	2.67	93.3	710	96.9	63.6
TS-1(4)	XG	Microwave (one step)	3.15	95.1	790	96.9	50.3
TS-1(5)	XG	Microwave (two steps)	2.41	75	260	96.4	63.3

[a] Conversion = (mol H_2O_2 consumed) × (mol H_2O_2 added)$^{-1}$.
[b] H_2O_2 selectivity = (mol epoxide obtained) × (mol H_2O_2 consumed)$^{-1}$.

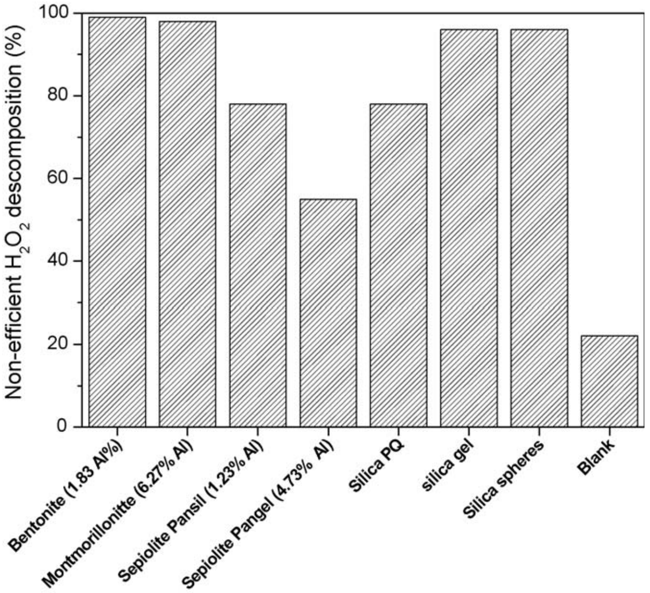

Fig. 4. Non-efficient decomposition of hydrogen peroxide using different inorganic binders.

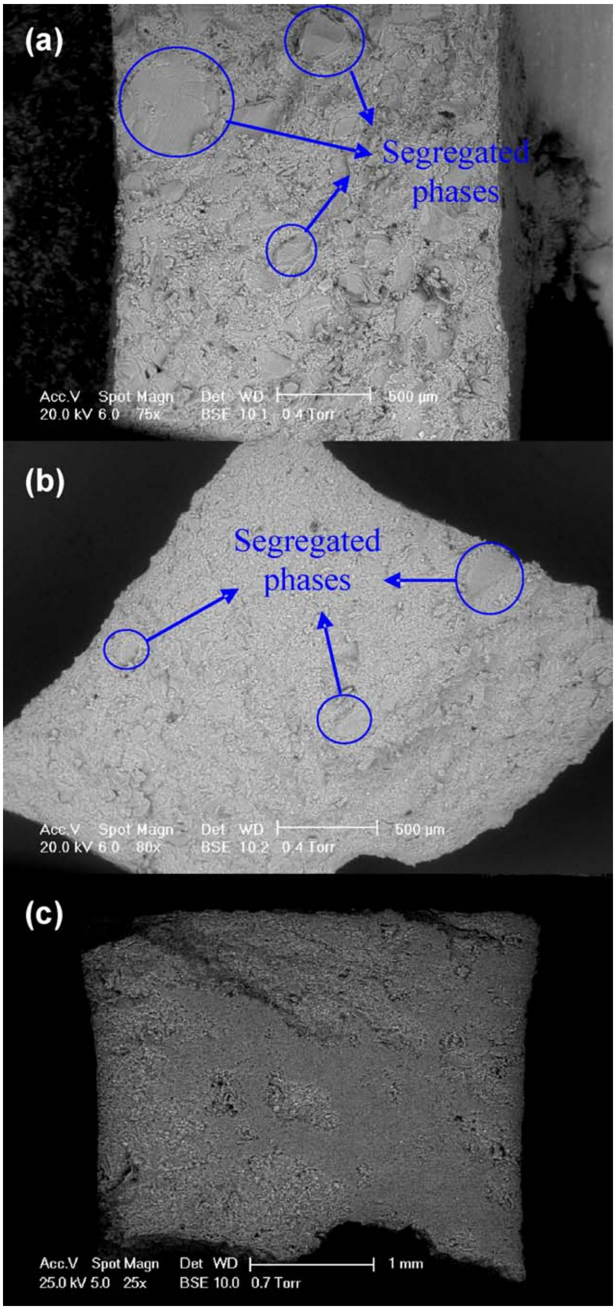

Fig. 5. SEM micrographs of extrudates obtained by different methods: (a) method 1, (b) method 2 and (c) method 3.

dry-mixing of the clay, methylcellulose and TS-1 powder with distilled water (200 wt% of solid). In method 2, the clay was added over a suspension of the TS-1 powder in water (200 wt% of solid) and method 3 was analogous to method 2 but using ultrasounds and larger water amount during the paste formation (240 wt% of solid). A TS-1 sample having a particle size comprising between 0.2 and 0.25 μm (measured by TEM) was employed as the catalytic component of the extrudate.

Morphological and surface properties of the final extrudates were determined by SEM (Fig. 5). It is clearly observed that the extrudates corresponding to methods 1 and 2 present rougher and more irregular surfaces, with many protuberances and hollows, than the extrudate obtained by method 3. For the former two samples, segregated phases with sizes between 100 and 250 μm can be detected, this fact being usually detrimental towards the mechanical strength of the extrudates. The surface of the extrudate prepared by method 3, however, is smoother with a higher degree of homogeneity and no phase segregation is perceptible. These observations are in agreement with this last sample exhibiting the highest physical strength (Table 2), which present physical strength values of 1.78 and 2.44 MPa, over the horizontal and vertical dimension of the particle, respectively. This better performance is attributed to the ultrasounds application over the aqueous suspension of TS-1 powder during the paste preparation. In method 3, ultrasounds prevent the aggregation of the zeolitic particles and, consequently, enhance their blending with the clay when the latter was added to the suspension, as it has been observed by SEM images. In addition, the crushing resistance of the extrudate prepared by method 3 was measured by Shell Test (SMS 1471), giving a maximum pressure value of 0.43 MPa, which can be considered appropriate for its use in fixed bed reactors.

Table 2
Influence of the extrusion method: mechanical strength of extruded material[a].

Method	Strength (MPa)	
	Horizontal	Vertical
1	1.27	1.58
2	1.58	2.37
3	1.78	2.44

[a] Measured over single particles.

3.4. Effect of the zeolite particle size

In order to evaluate the influence of the zeolite particle size in the mechanical strength of the extruded materials, three extruded samples were prepared using TS-1 zeolite samples with different particle sizes comprising between 0.2 and 1.5 μm (determined by SEM and TEM). These samples were extruded employing method 3, previously selected as the most suitable for the extrudates preparation, and weight proportions of TS-1/sepiolite of 70%/30%. The physical strength of these extruded materials was measured over individual particles and the resultant values are

Table 3
Influence of TS-1 particle size: mechanical strength of extruded material[a].

Particle size (μm)	Strength (MPa)	
	Horizontal	Vertical
0.2	1.48	2.44
0.7	1.10	1.88
1.5	1.07	1.33

[a] Measured over single particles.

shown in Table 3. It can be observed that the mechanical strength of the extruded catalyst increases as the zeolite particle size is smaller since this leads to a better blending and, therefore, a higher homogeneity of the paste during the extrusion process. These results indicate that the zeolite particle size has a significant effect on the mechanical properties of the extruded catalyst.

3.5. *Effect of the zeolite/binder ratio*

Once method 3 has been selected as the most suitable extrusion method, the effect of the binder amount in the paste was studied subsequently. Different extruded materials were obtained by preparing pastes with weight proportions of TS-1/sepiolite varying from 90%/10% to 50%/50%. For this study, the active phase used to prepare the extrudate was TS-1(5). Nevertheless, the large sample amount required for the extrusion process made necessary to scale up the synthesis procedure. In order to get larger zeolite quantities while keeping good crystalline and catalytic properties, it was necessary to increase the time of the two-stage microwave-heating program from 18 h/3.5 h to 20 h/8 h, respectively. XRD pattern and DR UV-Vis spectra of the resultant sample are presented in Fig. 6. The XRD spectrum confirms this new sample presents a high crystallinity, whereas the second technique corroborated the existence of Ti atoms into tetrahedral positions of the zeolitic framework, no extra-framework TiO_2 being detected. The titanium content of this sample, expressed as TiO_2, was 1.73 wt% and the synthesis yield obtained was 73%, producing a total amount of calcined sample of 90 g. As a consequence of the longer synthesis time applied, larger particle sizes were produced, achieving values in the range of 0.5–0.7 μm, which is in contrast with the mean value of about 260 nm obtained when the zeolite is prepared using the small scale system. These larger particle sizes could affect negatively the physical strength of the extrudates, due to a more difficult blending with the inorganic binder. The resultant mechanical strength values of the final extrudates, prepared using different zeolite/binder ratios, are presented in Table 4. It can be observed that the strength of the extrudates increases when the amount of the inorganic binder is higher, this increase being more pronounced when the sepiolite percentage is raised up to 40%. However, inorganic binder amounts higher than 40% lead to materials with very low Ti content, which would probably exhibit poor catalytic activity. Therefore, it was selected an optimal amount of inorganic binder of 40 wt%.

The reproducibility of the extrusion method employed was evaluated through the manufacture of three different sets of extruded catalyst, using weight proportions TS-1/sepiolite of 60%/40% and the previous TS-1 zeolite powder obtained in the large scale synthesis. The physical strength of these last sets was evaluated (over one particle) and the resultant values are presented in Table 5. The physical strength values obtained were very similar in all cases, indicating a high reproducibility of the extrusion method employed.

The catalytic activity of the extrudate containing 40% of inorganic binder was tested for propylene epoxidation in a fixed bed reactor using H_2O_2 as oxidant, loading 32 g of the extruded catalyst into the reactor. Fig. 7 illustrates the evolution of the catalytic activity and H_2O_2 selectivity as a function of time. Under these conditions, the H_2O_2 conversion and selectivity were close to 97% and 80%, respectively. The H_2O_2 selectivity to organic by-products such as propylene glycol and monomethyl ethers was

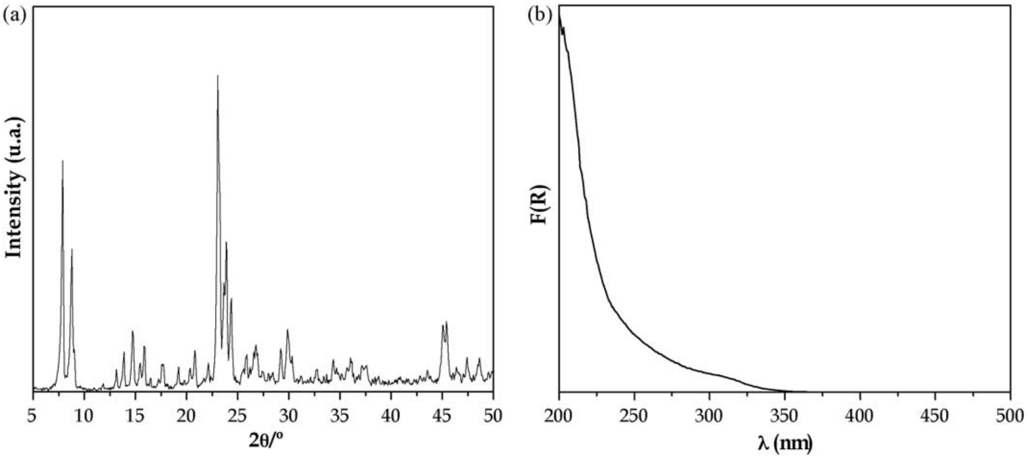

Fig. 6. (a) XRD patterns and (b) DR UV-Vis spectra of TS-1 sample prepared in large scale.

Table 4
Mechanical strength of extruded material as a function of the inorganic binder amount.

	Weight TS-1/sepiolite ratio (%/%)				
	90/10	80/20	70/30	60/40	50/50
Strength (MPa), horizontal	0.50	0.99	1.10	2.07	2.33
Strength (MPa), vertical	0.94	1.63	1.88	2.90	3.11

Table 5
Reproducibility of the extruded materials: mechanical strength.

	Weight TS-1/sepiolite ratio (60%/40%)		
	Set A	Set B	Set C
Strength (MPa), horizontal	2.12	2.09	2.11
Strength (MPa), vertical	2.81	2.89	2.91

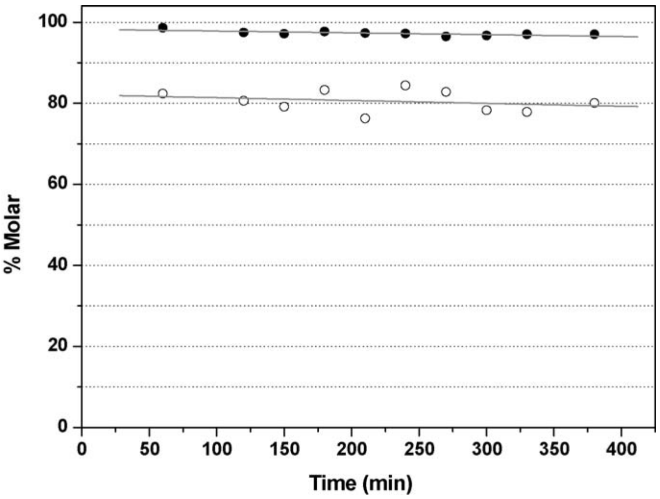

Fig. 7. Catalytic activity of the TS-1 extruded catalyst in propylene epoxidation with H_2O_2. (●) %H_2O_2 conversion and (○) %H_2O_2 selectivity.

very low, less than 5%. Accordingly, the selectivity to molecular oxygen in the reaction conditions tested were close to 15–17%.

Although a slight decrease in both activity and selectivity takes place during the reaction time tested, the catalyst exhibits significant stability, which is a remarkable and promising result regarding the potential application of this system at commercial scale. These results indicate that the binder type and amount, as well as the extrusion method, here optimized, allow TS-1 zeolite pellets to be obtained exhibiting good mechanical properties and convenient catalytic properties in olefin epoxidation reactions.

4. Conclusions

A method for the extrusion of TS-1 zeolite has been successfully developed for its application in propylene epoxidation. The best results were obtained when employing sepiolite and methylcellulose as inorganic and organic binders, respectively. The physical strength of the extruded materials depends on the zeolite particle size and the inorganic binder amount. It has been proved that the homogeneity of the extrusion paste depends on the zeolite particle size and the extrusion method used. The optimal extruded catalyst was prepared employing ultrasounds during the paste preparation, which avoids the formation of aggregates, and using weight proportions of TS-1/sepiolite of 60%/40%. Moreover, this method leads to catalysts with convenient mechanical strength for its use in a fixed bed reactor. The catalytic activity of this extruded TS-1 zeolite was evaluated for the propylene epoxidation reaction in a fixed bed reactor using H_2O_2 as oxidizing agent, achieving both high conversion and selectivity, with values close to 97% and 80%, respectively, which just decreased slightly along the reaction time.

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