SYNTHESIS OF PURE SILICA ZEOLITES WITH LOW CONTENT IN ORGANIC STRUCTURE DIRECTING AGENTS

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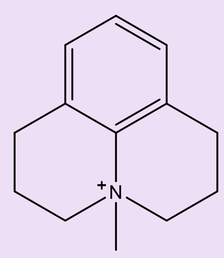
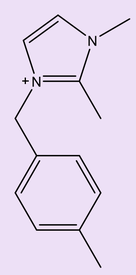
Objective

Synthesis of pure silica zeolites by using the minimum amount of organic structure directing agents (OSDA) with the objective of reducing the prize of the preparation.

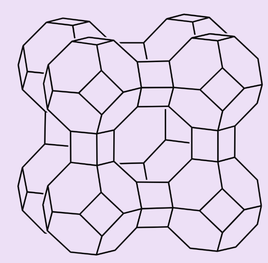
This study is focused on the pure silica LTA zeolite (**ITQ-29**). In this work, we have made a comparison between two synthesis procedures[1,2] using different OSDAs (Figures 1 and 2) and seeds to promote the crystallization



OSDA 1



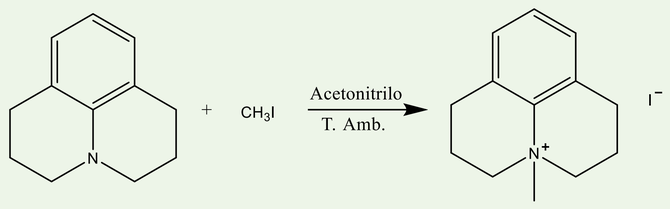
OSDA 2



LTA estructure

Synthesis (Method I)

The OSDA was prepared by alkylation reaction of julolidine with methyl iodide. Finally, the iodide salt was converted into the hydroxide form by ion exchange with a resin.



In the standard zeolite synthesis, the molar composition of the gel is:

SiO_2 : 0.25 R(OH) : 0.25 TMAOH : 0.5 HF : 3 H_2O

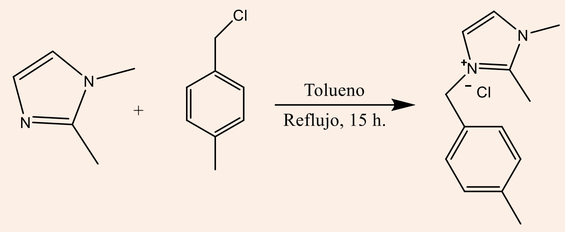
The mixture was crystallized at 448 K under rotation. It is necessary to use seeds to promote the crystallization of the material.

OSDA (%)	t (days)	Seed (%)
100	5	5
80	3	10
60	3	10
40	3	10

When the amount of OSDA (R) is decreased, the amount of seeds has to be increased.

Synthesis (Method II)

The OSDA was prepared by alkylation reaction of 1,2 dimethylimidazole with 4 methylbenzyl chloride. Finally, the iodide salt was converted into the hydroxide form by ion exchange with a resin.



In the standard zeolite synthesis, the molar composition of the gel is:

SiO_2 : 0.45 R(OH) : 0.05 TMAOH : 0.5 HF : 5 H_2O

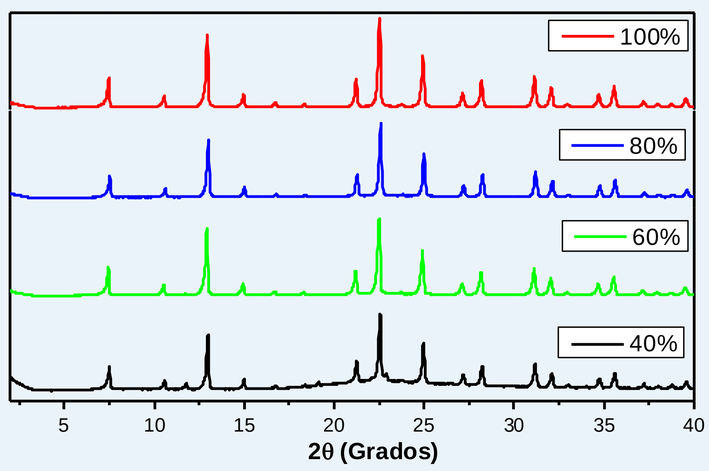
The mixture was crystallized at 448 K under rotation. It is necessary to use seeds to promote the crystallization of the material.

When the amount of OSDA (R) is decreased, it is necessary to use seeds.

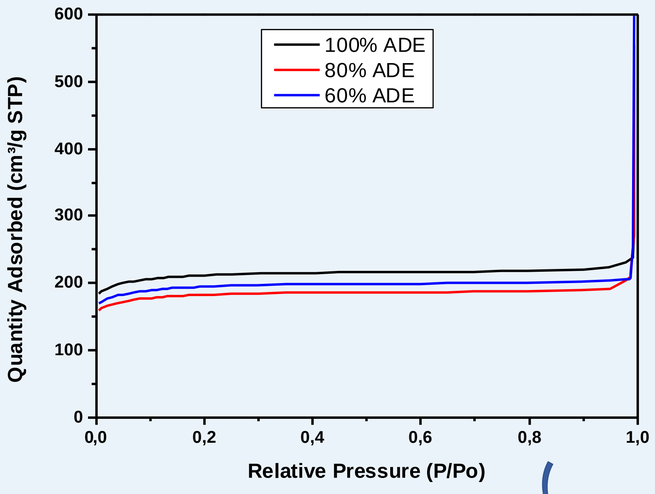
OSDA (%)	t (days)	Seed (%)
100	7	-
55	7	10
44	7	10
22	7	10

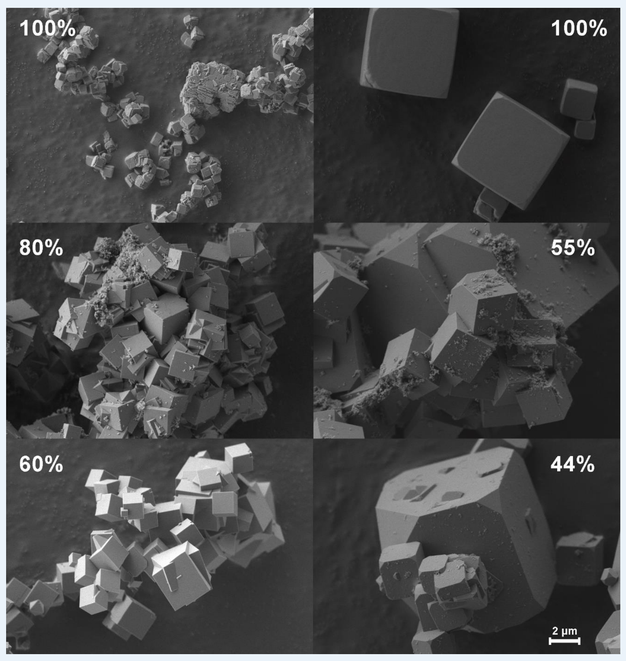
Characterization

Elemental analysis and ^{13}C MAS NMR (not shown) indicate that the OSDA remains intact after the synthesis. The organic can be removed by calcination in air. XRD patterns show that the crystallinity is maintained upon decreasing the amount of OSDA1 by 60% and OSDA2 by 44%.



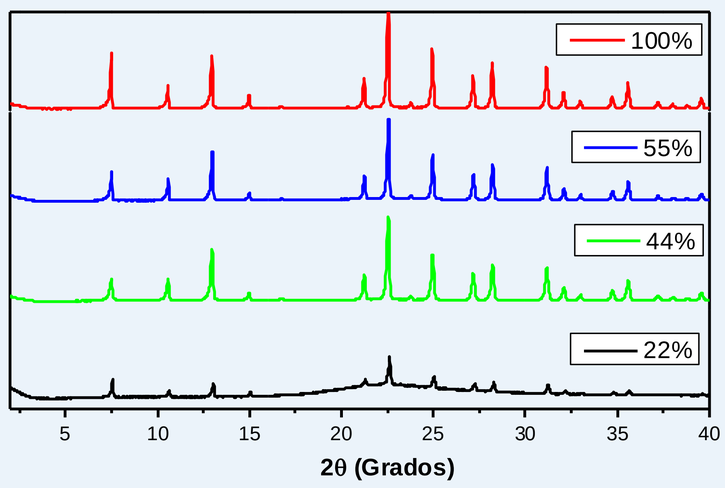
N_2 isotherms at 77K.



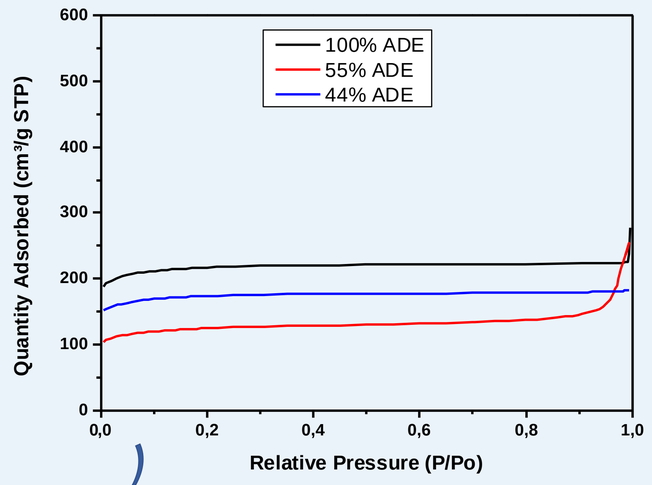


FESEM shows the presence of cubical crystals.

OSDA 1 → **Crystals of 2 µm**
OSDA 2 → **Crystals of 10 µm**



N_2 isotherms at 77K.



	% ADE	BET Area (m^2/g)	Micropore volume(cm^3/g)
Method I	100	692	0.33
	60	637	0.30
Method II	100	707	0.34
	44	556	0.27

Conclusions

The results obtained allow us to conclude that **it is possible to significantly reduce the amount of OSDA needed in the synthesis of pure silica ITQ-29 zeolite.**

OSDA 1 → **Decrease of 60 %**
OSDA 2 → **Decrease of 44 %**

Acknowledgments

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References

[1] A. Corma, F. Rey, J. Rius, M. J. Sabater, S. Valencia, *Nature.* **2004**, *431*, 287-290.
[2] B. W. Boal, et al, *Chem. Mater.* **2015**, *27*, 7774-7779.

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