Formation of Mullite and Spinel phases from SiO_2-Al_2O_3 gels prepared by spray pyrolysis technique. A ^{29}Si and ^{27}Al MAS-NMR study.

V. Diez, T. Gonzalez-Carreño, I. Sobrados, J. Sanz, S. Martinez.

Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid. España.

^{29}Si and ^{27}Al MAS-NMR techniques are very useful to analyze structural changes produced during thermal treatments of amorphous $SiO_2.Al_2O_3$ gels, with the aim of studying the mullite formation mechanism and to determine the compositional range in which mullite is formed.

In this work we have studied monophasic alumino-silicate gels prepared by spray pyrolysis, with Al/Si ratios between 1.5 to 18. NMR spectroscopy have been used to monitor cation environments and polyhedra condensation at increasing temperatures. In samples heated at 800º C, tetra-, penta- and octahedral aluminium were detected. In samples heated at 900º C, Al-rich mullite (Al/Si = 4-5) or Si-spinel nuclei (Al/Si = 10-18) are formed after the partial segregation of SiO_2 from alumino-silicate gels. The strong rearrangement of aluminium, with formation of tetra- and octahedral Al at expenses of pentahedral Al, is responsible for the exothermic peak detected at ~980º C in ATD curves. Thermal treatments between 1000 and 1200º C, produce the progressive incorporation of SiO_2 into mullite formed nuclei. In Al-rich samples, with nominal Al/Si between 8-18, the formation of mullite nuclei is only detected above 1100º C. In this case, spinel and mullite phases coexist in the temperature range 1100-1200º C.

Recently, we have prepared gels with variable compositions in the system SiO_2. Al_2O_3. B_2O_3 in order to incorporate boron atoms in mullite framework.

References

- Kanzaki, S.; Tabata, H.; Kumazawa, T.; Ohta, S., *J. Am. Ceram. Soc.*, **1985**, *68*, C6.
- Brown, I.W.N. MacKenzie, K.J.D.; Bowden, M.E.; Meinhold, R.H., *J. Am. Ceram. Soc*, **1985**, *68*, 298.
-Sanz, J.; Madani, A.; Serratosa, J.M.; Moya, S.; de Aza, S., *J. Am. Ceram. Soc.*, **1988**, *71*, C418.
- Sanz, J.; Sobrados, I.; Cavalieri, A.J.; Pena, P.; de Aza, S.; Moya, J., *J. Am. Ceram. Soc.*, **1991**, *74*, 2398.
- Schneider, H.; Merwin, L.; Sebald A.; *J. Mater. Sci.*, **1992**, *27*, 805.
- Gerardin, C.; Sundaresan, S.; Benziger, J.; Navrotsky, A., *Chem. Mat.*, **1994**, *6*, 160.
- Gonzalez-Carreño, T.; Sobrados, I.; Sanz, J., *Chem. Mat.*, **2007**, *19* (15), 3694.