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Structural characterization of siloxane-POE hybrid matrix as controlled drug delivery devices

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Abstract – The structural characterization of siloxane-POE hybrid matrix, prepared with polymer molecular weight 500 e 1900 g mol^{-1}, to be used as controlled drug delivery devices was done by ^{29}Si NMR, DSC and RXPD. Both the crystallinity and the gel formation, promoted by hydrolysis and condensation reactions, are dependent on the nature of the catalyst, the molecular weight of the polymer chain and the quantity of drug inside the matrix. The high polycondensation degree of hybrid matrix indicates the low number of terminal OH groups at this material structure.

The preparation of organic-inorganic hybrid materials precursors is a fundamental step to obtain gels from the sol-gel route. In this work, precursors were prepared by reacting the modified alkoxide , 3-(isocyanatopropyl) triethoxysilane (IsoTrEOS) with the modified polymer O,O'-bis(2-aminopropyl)poly-(propylene oxide), in tetrahydrofuran (THF) under reflux (at 80ºC for 24 h) [1] for polymers of molecular weight 500 e 1900 g mol^{-1}. The hydrolysis and polycondensation reactions were promoted by the addition of ethanol/water mixture containing HCl or NH_4F catalyst in the simple mixture of the non-hydrolyzed siloxane-PEO with the sodium diclofenac model drug giving rise to gelation. As a consequence of the nanostructural uniformity of the hybrid system transparent, flexible and a variety of forms of the xerogels were obtained after drying samples under vacuum at 70°C (Fig. 1). The structural characterization of these hybrid matrixes was done by ^{29}Si NMR, DSC and RXPD. The degree of crystallinity was determined from DSC data, using the ratio between the fusion enthalpy of the crystalline matrix and that of the 100% crystalline POE. For samples prepared with the high polymer length chain (POE1900) a significant increase of matrix crystallization was observed for samples containing low drug loading (1,0 % w/w), while that containing high amounts of drug show a drastic decrease of crystallinity. For samples prepared with the POE500 no endothermic peak was observed, indicating the matrix amorphization irrespective the drug concentration. The evolution of the XRPD patterns for samples containing increasing drug concentration (Fig. 2) corroborates the DSC results, that is, for POE1900, samples containing low drug loading show tha appearing of peaks indicating the crystallinity increases. The formation of O-Si-O bonds during polycondensation reactions was followed by ^{29}Si NMR spectroscopy, taking account for the presence of different silicon polyhedral species, T_n, where T designates the trifunctional Si and n the number of neighboring Si bonded by first coordination spheres oxygen atoms to the next Si. Irrespectively the molecular weight of the polymer of hybrid matrixes, only peaks corresponding to T_2 ($RSi(OSi)_2(OH)$) and T_3 ($RSi(OSi)_3$) species were observed. Note that the polycondensation degree was alleyways higher for samples prepared from the neutral catalyst, excepting for the one prepared with POE1900 containing the higher drug loading (17,0 %w/w). This set of results show that both the crystallinity and the gels formation are dependent on the catalyst nature, the molecular weight of the polymer moieties and on the amount of drug inside the matrix. Moreover, the high polycondensation degree of these matrixes reveals the presence of low quantity of OH terminals limiting the interactions drug matrix to two sites: the carbonyl of the urea group and the ether-type oxygen.

Figure 1: Different forms of siloxane POE1900 hybrid.

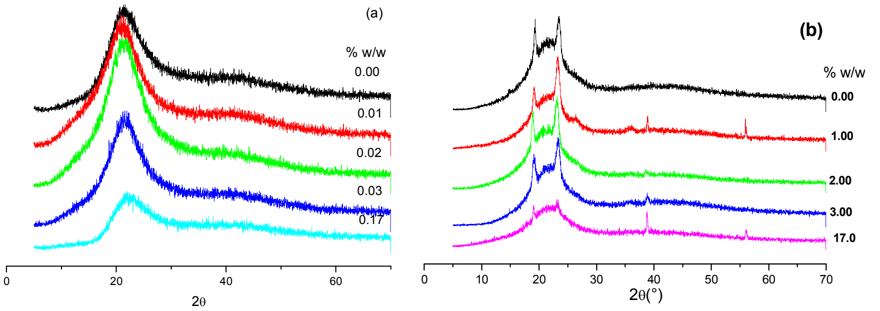

Figure 2: X-Ray Powder Diffraction Patterns of the hybrid matrixes a) POE 500 and b) POE 1900 as a function of the drug loading.

References

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