

## COMPATIBILITY OF POLYMERIC BINDERS AND POTENTIAL PLASTICISERS

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Plasticization of polymeric binding agents may enhance formation of coherent tablets from poorly compactable materials (Wells et al 1982). The ability of a molecule to plasticize a polymer is due to the compatibility of the mix and the resultant increase in mobility of the the polymer chains (Osterwald 1982). A polymer-plasticizer mix existing as a single phase can be regarded as a solution irrespective of its mechanical properties. Thermodynamic criteria for solution (i.e. a negative free energy change) can thus be applied to the problem of solid state compatibility. The entropy of mixing is low for polymer systems compared with non-polymers and therefore the heat of mixing determines the magnitude and sign of the free energy change.

The heat of mixing will be zero and compatibility assured by the entropy factor if the solubility parameter ( $\delta$ ) of both polymer and potential plasticizer are the same (Hildebrand and Scott 1950). We have adopted this approach to investigate compatibility between some tablet binders and potential plasticizers. The binders used were HPMC (Methocel E5), PVP (K25 and K90) and Starch 1500; potential plasticizers included a range of aqueous miscible monomeric glycols and PEGs 200 to 6000. Solubility parameter values for the polymers were determined using the solvent spectra method (Burrell 1975). Values for potential plasticizers were taken from the literature.

An alternative approach is that of Flory and Huggins (see Flory 1953). They derived an expression for the free energy change on mixing by inclusion of a semi-empirical heat of mixing term  $\chi$ . This term (the Flory-Huggins interaction parameter) characterises the degree of interaction between polymer and diluent. Where  $\chi$  exceeds a critical value, dependent on molar volumes, immiscibility is observed. If  $\chi$  is less than the limiting value of 0.5 compatibility in all proportions is assured. Between the two extreme cases partial compatibility occurs. We have estimated values for  $\chi$  from solubility parameters, plasticizer molar volumes and an assumed value of 0.4 for the entropy constant.

Starch 1500 showed minimal solubility in the test solvents, hence values for  $\delta$  and  $\chi$  could not be determined. This is ascribed to the crystalline structure of starches. Plasticization of such materials is generally precluded.

Solubility parameter limits for HPMC in strongly hydrogen bonded solvents were  $23.15 \pm 2.45$  ( $\text{Jm}^{-3}$ )<sup>0.5</sup>. No plasticizer screened was thermodynamically ideal for HPMC. Although the PEGs have  $\delta$  values of  $24.55 \pm 5.15$  ( $\text{Jm}^{-3}$ )<sup>0.5</sup> i.e. apparently within the limits for compatibility with HPMC, values for  $\chi$  were 0.54 for PEG 200 increasing with molecular weight to 2.19 for PEG 6000 indicating that only partial compatibility could be expected.

In contrast the PVP grades ( $\delta = 24.56 \pm 5.13$  ( $\text{Jm}^{-3}$ )<sup>0.5</sup>) should be compatible in all proportions with PEGs, since the  $\delta$  values for the PEGs approach those of the PVPs. For low molecular weight liquid PEGs  $\chi$  values were close to 0.4. PEGs 1500 to 6000 had  $\chi$  values in the range 0.45 to 0.58. Propylene glycol ( $\delta = 25.8$  ( $\text{Jm}^{-3}$ )<sup>0.5</sup>,  $\chi = 0.45$ ) should also be compatible with the PVPs.

Visual assessment of the extent of compatibility in aqueous cast films was in general agreement with the above theoretical predictions. Limited compatibility observed with PEGs 1500 to 6000 and the binders was due to PEG crystallinity and low entropy of mixing for polymers.

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