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Multiscale structure in amorphous carbohydrate matrices in relation to phase behavior, molecular packing and interaction with water

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Glassy matrices for the encapsulation and stabilization of bioactive compounds often consist of molecularly miscible blends of a carbohydrate of higher molecular weight and a low molecular weight carbohydrate or polyol. Typical examples of such blends are maltodextrin/sucrose and trehalose/glycerol. The optimization of the barrier properties of such matrices depends in a subtle way on the balance between the plasticization and antiplasticization of the high molecular weight carbohydrate by the low molecular weight compound [1]. Here, we report on the phase behavior of encapsulation matrices consisting of blends of a hydrophobically-modified starch and sucrose. By differential scanning calorimetry (DSC), it is shown that the blends exhibit two glass transitions, with the specific heat and the temperatures of the transitions varying in a systematic way with the blend composition [2]. This hints at a limited degree of phase separation of the matrix into two amorphous phases. We present a model to calculate the degree of phase separation of the blends and the composition of the phases from the dependence of the glass transition temperatures of the blends on the sucrose content. The antiplasticization of the starch-rich phase by sucrose is confirmed by the decrease in molecular hole size with increasing sucrose content as determined by positron annihilation lifetime spectroscopy (PALS) [3] and the stiffening of the starch chains solid state NMR by the addition of sucrose to the matrix. We conclude the lecture by discussing the advantages and disadvantages of the two-phase system for its use in encapsulation systems.

References

- [1] J. Ubbink, *Advanced Drug Delivery Reviews* **100**, 10-26 (2016).
- [2] C. Tedeschi, B. Leuenberger and J. Ubbink, *Food Hydrocolloids* **58**, 75-88 (2016).
- [3] D. Hughes, C. Tedeschi, B. Leuenberger, M. Roussanova, A. Coveney, R. Richardson, G. Badolato Bönisch, M.A. Alam and J. Ubbink, *Food Hydrocolloids* **58**, 316-323 (2016).

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