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Collagenous Bioadhesives: Structure-Property Correlations and Hygrothermal Ageing

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Collagen-based bioadhesives, also known as animal glues, are derived from animal parts (e.g. skin, bone, and cartilage). They have been widely used in the production of decorative wood working, paintings, bookbinding, and other historical objects for centuries as well as for their restoration and preservation [1].

Collagen, in which the term (kólla) is derived from the Greek word for glue, is a fibrous structural protein with a hierarchical microstructure at different length scales. Collagen is water insoluble and can be turned into water-soluble gelatine through the denaturation process. Upon denaturation, the triple helix structure in collagen unwinds and turns into random coils giving gelatinous animal glue. Gelatinous animal glue can be dissolved in water and during gelling and subsequent drying, the random protein coils undergo partial renaturation back into triple helices. These triple helices in the adhesive act as physical cross-links leading to a continuous three-dimensional network structure [2]. These microstructural features affect the physical and mechanical performance of the glue at micro and macro scales.

In this study four different adhesives from different animal species both mammalian and fish are investigated. These adhesives are bovine bone, bovine skin, rabbit skin, and fish. For this study, thin films of these adhesives were manufactured using the solution casting method. Physical and mechanical behaviour of the adhesives were characterized using Differential Scanning Calorimetry (DSC), X-ray diffraction (XRD), Bloom strength evaluation, and uniaxial tensile tests.

This study finds linear correlations between microstructure and macroscopic performance of the adhesives. Particularly, the results demonstrate a strong correlation between the Bloom strength and the amount of microstructural features in the form of triple helices measured by both DSC and XRD techniques (see figure 1). Moreover, uniaxial tensile tests on the adhesive films show a strong correlation between the level of renaturation of triple helices in the adhesive structure with the toughness of the adhesives. However, the stiffness is hardly influenced. Fish glue is identified as the toughest adhesive amongst the tested glues, whilst bovine bone glue is shown to be the more brittle adhesive and the weakest. Moreover, hygrothermal accelerated ageing of the adhesives films was performed by applying raised temperature and cyclic relative humidity. The changes of microstructural features as a function of ageing were measured by DSC and XRD. The results demonstrated a reduction of the triple helix content as an ageing pathway in these adhesives. Moreover, the gradual reduction of triple helices in the adhesives was accompanied by loss of tensile toughness. Bovine bone glue was found to be the most susceptible to hygrothermal ageing.

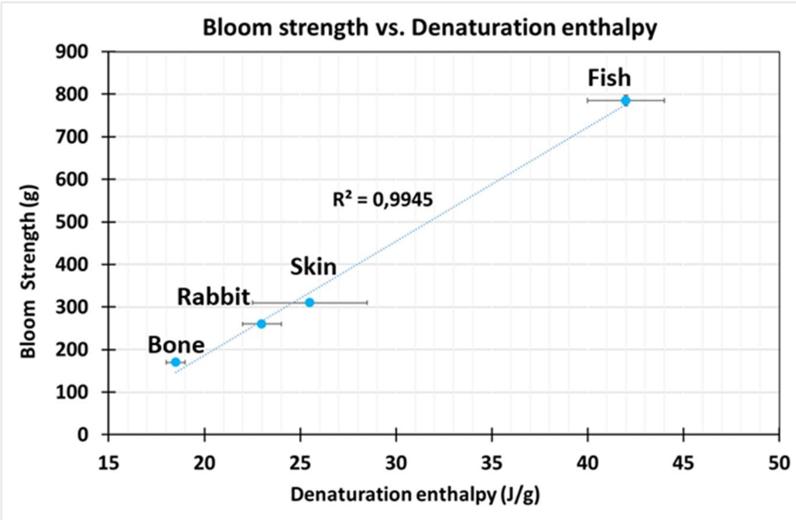

Figure 1. A linear correlation between denaturation enthalpy obtained by DSC and Bloom (gel) strength for four different adhesives.

References

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