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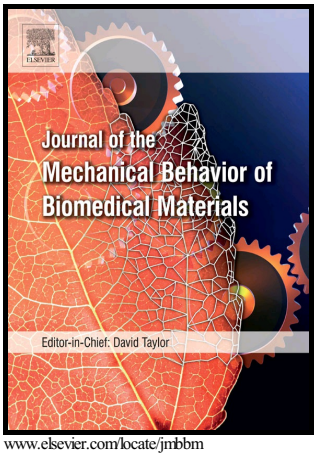

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Formulation and viscoelasticity of mineralised hydrogels for use in bone-cartilage interfacial reconstruction

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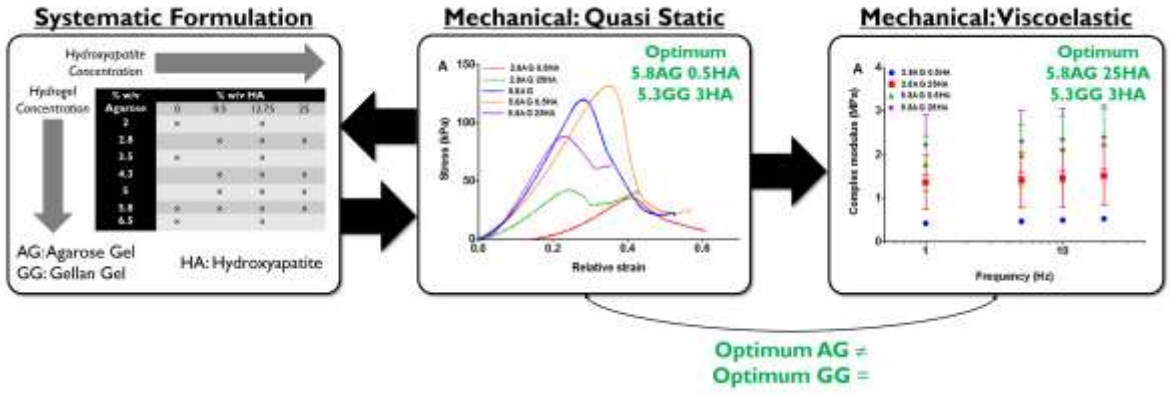
Abstract
Articular cartilage is a viscoelastic tissue whose structural integrity is important in maintaining joint health. To restore the functionality of osteoarthritic joints it is vital that regenerative strategies mimic the dynamic loading response of cartilage and bone. Here, a rotating simplex model was employed to optimise the composition of agarose and gellan hydrogel constructs structured with hydroxyapatite (HA) with the aim of obtaining composites mechanically comparable to human cartilage in terms of their ability to dissipate energy. Addition of ceramic particles was found to reinforce both matrices up to a critical concentration (<3 w/v%). Beyond this, larger agglomerates were formed, as evidenced by micro computed tomography data, which acted as stress risers and reduced the ability of composites to dissipate energy demonstrated by a reduction in tan δ values. A maximum compressive modulus of 450.7±24.9 kPa was achieved with a composition of 5.8 w/v% agarose and 0.5 w/v% HA. Interestingly, when loaded dynamically (1 – 20 Hz) this optimised formulation did not exhibit the highest complex modulus instead a sample with a higher concentration of mineral was identified (5.8 w/v% agarose and 25 w/v% HA). Thus, demonstrating the importance of examining the mechanical behaviour of biomaterials under conditions representative of physiological environments. While the complex moduli of the optimised gellan (1.0±0.2 MPa at 1 Hz) and agarose (1.7±0.2 MPa at 1 Hz) constructs did not match the complex moduli of healthy human cartilage samples (26.3±6.5 MPa at 1 Hz), similar tan δ values were observed between 1 and 5 Hz. This is promising since these frequencies represent the typical heel strike time of the general population. In summary, this study demonstrates the importance of considering more than just the strength of biomaterials since tissues like cartilage play a more complex role.

Graphical Abstract



Keywords; Hydrogels, cartilage, viscoelastic behaviour, dynamic mechanical analysis, hydroxyapatite