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Hydration dependent mechanical performance of denture adhesive hydrogels

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ABSTRACT

Objective. Hydration in denture adhesives regulates the formation of complex morphologies and mechanical function. Multiscale experimental approaches are required to evaluate the impact of hydration on the inherent heterogeneity of denture adhesive-based hydrogels at different length scales and the impact of such phenomena on adhesion performance.

Methods. The morphology of hydrated denture adhesives was examined via cryo-scanning electron microscopy (cryo-SEM). The rheological and thermodynamic behaviour of bulk hydrated deture adhesives was examined by rheology and differential scanning Calorimetry (DSC). The microscopic mechanical properties of the denture adhesives were characterised by atomic force microscopy (AFM) and compared to the properties measured at the macroscopic scale.

Results. The rheological and mechanical properties of commerically available denture adhesive hydrogels were found to be critically dependent on both the formulation of the adhesives and their hydration level. Clear progression of phase separation was observed in hydrated denture adhesives as hydration increased and changed the mechanical properties of the adhesives at multiple length scales. The adhesives displaying more heterogeneous structures, which were associated with the presence of hydrophobic and organic compounds in the formulation, exhibited more variable mechanical behaviour and weaker rheological properties, but stronger adhesive properties.

Significance. Our results are important in defining the relationships between hydrophilicity, hydration, mechanical and adhesive properties of denture adhesives, allowing the development of improved chemical formulations that control the fixation of dentures.

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1. Introduction

Denture adhesives are commonly used to promote denture retention, stability and function in oral applications. These materials are usually based on macromolecules that can sustain the formation of numerous hydrogen bonds and contribute to the strong non-covalent bonding once mixed with mucus and saliva, to acquire viscous and adhesive properties [1]. Most of these adhesives contain both hydrophobic and hydrophilic molecules, which are designed to balance their bonding strengths to the gum and the denture [2–5]. Understanding the mechanical behaviour of the hydrated adhesives is required for the design of improved bioadhesives. Specifically, relationships between adhesive mechanical performance, formulation and hydration of denture adhesive are needed, in order to improve our control of the fixation of dentures. Our previous study highlighted marked differences in failure mechanisms of commercial denture adhesives (PDFP and PDAC) [6]. The existence of hydrophobic, organic compounds in the formulation of PDAC and the associated inherent heterogeneity of the resulting materials was proposed to be a critical factor responsible for the variation of bulk adhesive performance.

Hydration within the oral cavity is proposed to contribute to generate a complex morphology affecting the mechanical function of denture adhesives. Bioadhesives can absorb considerable amounts of water due to their hydrophilicity and therefore function is sensitive to hydration. From fundamental considerations, the hydration of the bioadhesives may support the formation of interlocked physical chain entanglement, electrostatic interactions and secondary chemical interactions (hydrogen bonding) that contribute to the bulk mechanical and rheological properties of the corresponding biomaterials [7–10]. Previous research also inferred that the hydrophilicity and solubility of some of the macromolecules within the adhesive composition could lead to leaching of molecules and reduce structural and functional properties of the adhesives [11]. Overly hydrophilic materials were found to lead to the fast deterioration of the mechanical properties of adhesives and to cause hydrolytic degradation [12–14]. However, a study of the role of hydration on the microstructure and mechanical properties of different types of denture adhesives is lacking. Therefore, a detailed understanding of the relationships between hydrophilicity, water uptake and mechanical and adhesive properties of the formulations currently used as denture adhesives is required [2,3,15], especially across a range of length scales shown to be critical in defining adhesive performance [6].

This work therefore aims to evaluate the impact of hydration on the inherent heterogeneity of denture adhesive-based hydrogels at different length scales and the impact of such phenomena on adhesion performance. The mechanical properties of hydrogel formulations used as denture adhesives, after hydration at different levels, were characterised via nanoscale indentation, by atomic force microscopy (AFM) [16–18]. The morphology of these hydrogels was visualised using scanning electron microscopy operating under sample cryogenic conditions (cryo-SEM). The influence of absorbed water and the level of hydration of the corresponding macromolecules was further quantified through differential scanning calorimetry (DSC).

Table 1 – Weight ratios applied to the denture adhesives (PDAC and PDFP) mixed with DI water.

Adhesives	Weight ratio of DI water					
PDAC	No water	10:1	1:1	1:2	1:4	1:10
PDFP	n/a	n/a	1:1	1:2	1:4	1:10

2. Materials and methods

2.1. Materials

Two commercial adhesives, Poligrip®, Ultra Wernets®, Denture Fixative Powder (PDFP) and Poligrip®, Ooze-Control Tip® Denture Adhesive Cream (PDAC) were studied in this work. These adhesives were selected because of their comparable compositions, but difference in types of formulation (PDFP is a powder whereas the PDAC is a cream). We made a mention of this in the introduction. PDFP powder is composed of poly(methylvinylether/maleic acid) sodium–calcium mixed partial salt, cellulose gum and aroma. PDAC cream consists of poly(methylvinylether/maleic acid) sodium-calcium mixed partial salt, petrolatum, cellulose gum, mineral oil, silica, poly(methylvinylether/mal2eic acid), flavour, Red 30 aluminium lake and Red 7 calcium lake. PDAC contains more hydrophobic compounds such as hydrocarbon vehicles (mineral oil and petrolatum) compared to PDFP, in addition to MVE/MA copolymer, which may affect the hydration of the polymers and gel formation resulting in different adhesion behaviour. In order to evaluate the influence of water added in the denture adhesive hydrogels, series of samples with different DI water weight ratios as illustrated in Table 1 were prepared. All the samples were examined immediately after the hydrogel was macroscopically homogeneous. For PDAC, due to its cream formulations, pure adhesive with no water mixed was examined first. Water was gradually added to the adhesives at a weight ratio of water to adhesives starting at 10:1, followed by higher water ratios of 1:1, 1:2, 1:4 and 1:10. For PDFP, only four water ratios (1:1, 1:2, 1:4 and 1:10) were prepared since pure PDFP is dry powder.

2.2. Cryo-SEM

Cryo-SEM is a low temperature electron microscopy technique, which involves the examination of materials below ambient temperature (typically between −100 to −170 °C) and allows the structure and morphology of the sample to be better preserved and imaged in a hydrated and chemically unaltered state. Cooling of the system is achieved with liquid nitrogen. The denture adhesives were first fixed on the cryo-stage using the optimal cutting temperature (OCT) compound glue. The cryo-stage with adhesive samples was then brought to dip into liquid nitrogen to satisfy cryo-temperature (−130 °C) and transferred in vacuum to the cold-stage of the cryo-prechamber of the SEM system (Quanta 3D FEG, EU/USA). The adhesive sample was then cut horizontally to create a cross-sectional area facing the electron detector using a sharp blade equipped within the prechamber. Afterwards, the tem-

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