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Multiscale modelling and diffraction-based characterization of elastic behaviour of human dentine

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ABSTRACT

Human dentine is a hierarchical mineralized tissue with a two-level composite structure, with tubules being the prominent structural feature at a microlevel, and collagen fibres decorated with hydroxyapatite (HAp) crystallite platelets dominating the nanoscale. Few studies have focused on this two-level structure of human dentine, where the response to mechanical loading is thought to be affected not only by the tubule volume fraction at the microscale, but also by the shape and orientation distribution of mineral crystallites, and their nanoscale spatial arrangement and alignment. In this paper, in situ elastic strain evolution within HAp in dentine subjected to uniaxial compressive loading along both longitudinal and transverse directions was characterized simultaneously by two synchrotron X-ray scattering techniques: small- and wide-angle X-ray scattering (SAXS and WAXS, respectively). WAXS allows the evaluation of the apparent modulus linking the external load to the internal HAp crystallite strain, while the nanoscale HAp distribution and arrangement can be quantified by SAXS. We proposed an improved multiscale Eshelby inclusion model that takes into account the two-level hierarchical structure, and validated it with a multidirectional experimental strain evaluation. The agreement between the simulation and measurement indicates that the multiscale hierarchical model developed here accurately reflects the structural arrangement and mechanical response of human dentine. This study benefits the comprehensive understanding of the mechanical behaviour of hierarchical biomaterials. The knowledge of the mechanical properties related to the hierarchical structure is essential for the understanding and predicting the effects of structural alterations that may occur due to disease or treatment on the performance of dental tissues and their artificial replacements.

1. Introduction

Dentine is a hydrated biological mineral composite tissue with a hierarchical structure and versatile mechanical properties [1]. At the microscopic level, dentine has a well-oriented structure with an arrangement of dentinal tubules that extend throughout the entire dentine thickness, from the amelo-dentinal junction (ADJ) to the pulp [1–3]. At the nanoscale, dentine is a composite of plate-like hydroxyapatite crystals (HAp) that have the shape of elongated pancakes (~2–4 nm thick, ~30 nm wide and up to 100 nm long) randomly embedded in a collagen matrix [4,5]. Characterizing the mechanical properties of the tissue according to its complex hierarchical structure benefits the understanding of the internal architecture and hierarchical properties of materials.

Previously, most research in this area has concentrated on the mechanical properties of dentine at the macro- and microscale, i.e. Young's modulus, Poisson's ratio, hardness and fracture properties, using a variety of measurement methods [6]. However, few studies have focused on the nanoscale, where mechanical alterations can be considered to be a function of crystal shape and orientation of the mineral phase [7,8]. During elastic loading, stresses are expected to be transferred to the stiff HAp platelets from the surrounding collagen matrix [7]. To investigate this, it is required to use techniques that allow in situ quantification of the mechanical response of nanoscale HAp phases to loading.

Synchrotron-based X-ray diffraction, and small- and wide-angle X-ray scattering (SAXS and WAXS, respectively), are advanced non-destructive techniques that enable characterization of the nanoscale and subnanoscale structure of materials, and have been widely used to study load transfer between two phases in matrix composites [9–11]. In situ compression testing, in combination with the WAXS technique, has been used to quantify the internal strains of

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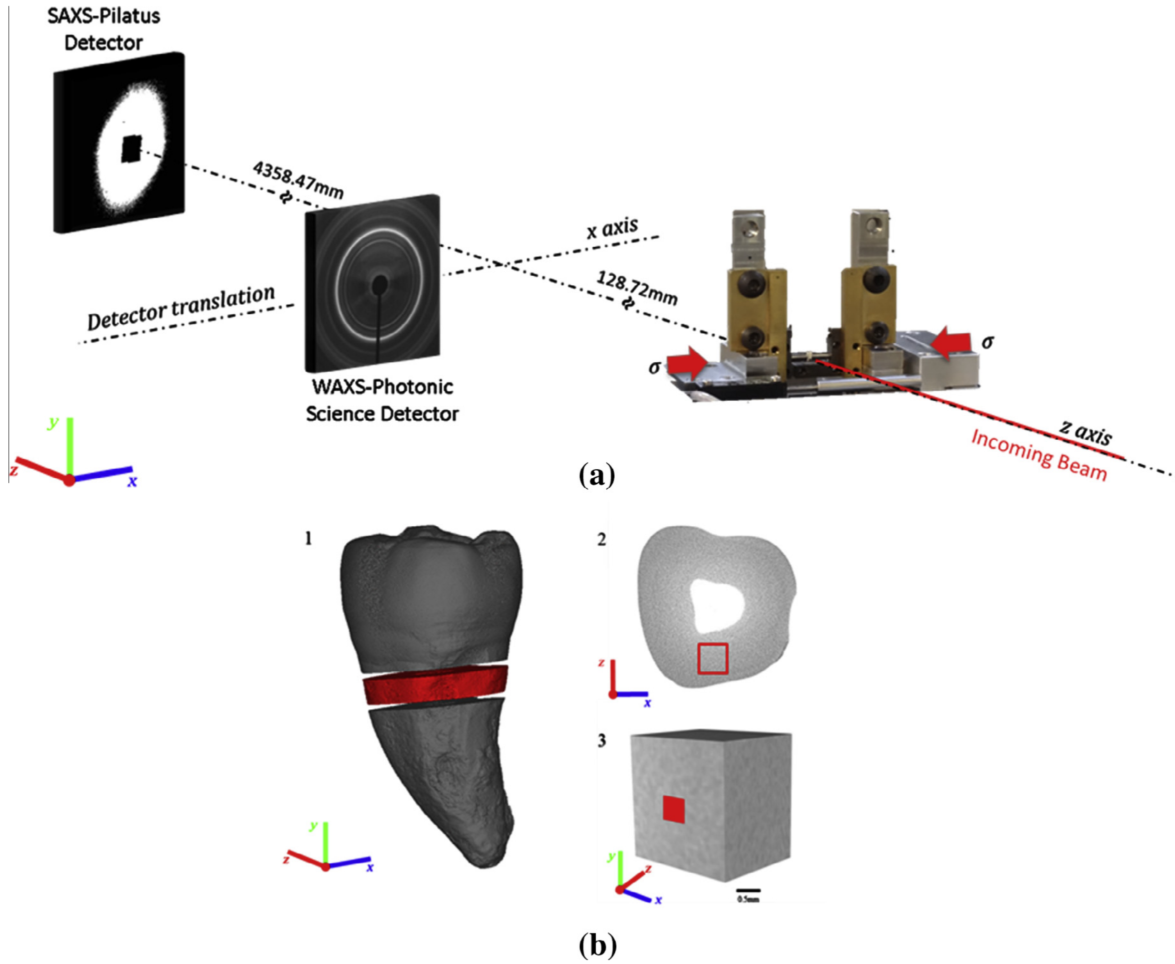

Fig. 1. Schematic diagram of experimental setup and sample preparation. (a) Sample under uniaxial compressive loading on the compression stage. The monochromatic X-ray beam was directed perpendicular to the sample surface and the loading direction. WAXS and SAXS diffraction patterns were recorded at each loading step at three locations on the sample. The WAXS detector was translated laterally out of the beam to expose the SAXS detector after each collection of WAXS. (b) Micro-CT-based models of the three preparation stages. (1) 2 mm thick dentine disk (coloured red) cut below the enamel–cement line and (2) further cut and polished to produce (3) the final 2 mm × 2 mm × 2 mm cubes of dentine. The red square indicates the central position of the X-ray beam.

the phase [9–11]. In addition, for mineralized tissues, SAXS is able to reveal quantitative nanoscale information about the structure, orientation and degree of alignment of crystals [12]. Those parameters have been identified as critical for the mechanical properties and stability of the materials [13–15]. It is only recently that this technique has been applied to the study of mechanical behaviour in mineralized biological composites such as bone [16–19] and teeth [7,8,20]. Deymier-Black et al. [7] determined the longitudinal apparent modulus of HAp in bovine dentine using synchrotron-based WAXS, while strain distribution across the ADJ in bovine teeth was investigated by Almer and Stock [20]. A deep understanding of the relationship between the nanoscale structure and macroscopic mechanical behaviour is lacking. Earlier studies do not take into account the nanoparticle distribution [7,20], which can be derived from SAXS data. In addition, all these studies were carried on non-human samples, in which different particularities of the tubule structure and morphology are expected to result in differences in the mechanical properties [21].

In parallel, various analytical models of composites have been proposed to describe the interaction of different mineral phases and model the elastic properties of hard tissues (e.g. [22–24]). Besides these, one widely accepted model is the Eshelby inclusion model [25,26]. Recently, the Eshelby model has also been applied in dental research based on nanoindentation and finite-element model data [27–30] to explain and predict the elastic response of dentine on the microscopic level. However, the models used in previous simulations were limited, in which no consideration was given to the nanoscale structure, and this led to discrepancies of overestimation between the predictions and experimental results [7].

In order to improve the understanding of the influence of the nanoscale structure variation of the two-level composite of human dentine on its mechanical response, in this study, the in situ synchrotron X-ray techniques (simultaneous SAXS/WAXS) were used to measure the elastic strain (WAXS), alterations in crystal orientation and degree of alignment of HAp phases (SAXS) in human dentine under externally uniaxial compressive loading along two directions, longitudinal as well as transverse with respect to the preferential tubule direction. Meanwhile, an extended multiscale Eshelby model for a two-level composite was established. The capability of the model in capturing the relationship between the nanoscale structure and macroscopic loading was evaluated.

2. Materials and methods

2.1. Sample preparation

Two freshly extracted sound human third molars (ethical approval obtained from the National Research Ethics Committee; NHS-REC reference 09.H0405.33/Consortium R&D No. 1465) were