Acta Biomaterialia 57 (2017) 373–383

Contents lists available at ScienceDirect

Acta Biomaterialia

journal homepage: www.elsevier.com/locate/actabiomat

Full length article

On the relationship between indentation hardness and modulus, and the damage resistance of biological materials

David Labonte [a],*, Anne-Kristin Lenz [b], Michelle L. Oyen [a]

[a] The NanoScience Centre, Department of Engineering, Cambridge, UK
[b] University of Applied Sciences, Bremen, Germany

ARTICLE INFO

Article history:
Received 17 February 2017
Received in revised form 20 April 2017
Accepted 15 May 2017
Available online 22 May 2017

Keywords:
Structure-function relationships
Biomechanics
Wear
Hydration

ABSTRACT

The remarkable mechanical performance of biological materials is based on intricate structure–function relationships. Nanoindentation has become the primary tool for characterising biological materials, as it allows to relate structural changes to variations in mechanical properties on small scales. However, the respective theoretical background and associated interpretation of the parameters measured *via* indentation derives largely from research on 'traditional' engineering materials such as metals or ceramics. Here, we discuss the functional relevance of indentation hardness in biological materials by presenting a meta-analysis of its relationship with indentation modulus. Across seven orders of magnitude, indentation hardness was directly proportional to indentation modulus. Using a lumped parameter model to deconvolute indentation hardness into components arising from reversible and irreversible deformation, we establish criteria which allow to interpret differences in indentation hardness across or within biological materials. The ratio between hardness and modulus arises as a key parameter, which is related to the ratio between irreversible and reversible deformation during indentation, the material's yield strength, and the resistance to irreversible deformation, a material property which represents the energy required to create a unit volume of purely irreversible deformation. Indentation hardness generally increases upon material dehydration, however to a larger extent than expected from accompanying changes in indentation modulus, indicating that water acts as a 'plasticiser'. A detailed discussion of the role of indentation hardness, modulus and toughness in damage control during sharp or blunt indentation yields comprehensive guidelines for a performance-based ranking of biological materials, and suggests that quasi-plastic deformation is a frequent yet poorly understood damage mode, highlighting an important area of future research.

Statement of Significance

Instrumented indentation is a widespread tool for characterising the mechanical properties of biological materials. Here, we show that the ratio between indentation hardness and modulus is approximately constant in biological materials. A simple elastic-plastic series deformation model is employed to rationalise part of this correlation, and criteria for a meaningful comparison of indentation hardness across biological materials are proposed. The ratio between indentation hardness and modulus emerges as the key parameter characterising the relative amount of irreversible deformation during indentation. Despite their comparatively high hardness to modulus ratio, biological materials are susceptible to quasi-plastic deformation, due to their high toughness: quasi-plastic deformation is hence hypothesised to be a frequent yet poorly understood phenomenon, highlighting an important area of future research.

1. Introduction

In search for inspiration for the design of novel materials with enhanced properties, biological materials receive an increasing amount of attention (e.g. [1–5]). Two features of these materials stand in stark contrast to 'classic' engineering materials: first, they are hierarchical in design, that is they are constructed from multiple building blocks arranged on characteristic length scales ranging from a couple of nanometres to a few millimetres. Second, the structural arrangement and properties of these components are tailored to specific functional demands.

---

* Corresponding author.
E-mail address: dl416@cam.ac.uk (D. Labonte).

http://dx.doi.org/10.1016/j.actbio.2017.05.034
1742-7061/

List of symbols

A_1—A_5	indentation constants
a_r	width of residual indent
a	contact radius
β	equivalent cone angle
c_f	characteristic flaw size
c	crack length
E'	plane strain modulus
E	Young's modulus
E_I	indentation modulus
$E_{I,d}$	indentation modulus measured in dehydrated conditions
$E_{I,w}$	indentation modulus measured in hydrated conditions
G_c	strain energy release rate
H_I	indentation hardness
$H_{I,d}$	indentation hardness measured in dehydrated conditions
$H_{I,w}$	indentation hardness measured in hydrated conditions
h_e	reversible indentation depth
h_{max}	maximum indentation depth
h_r	residual (irreversible) indentation depth
h	indentation depth
I_E	elasticity index
R_{ID}	resistance to irreversible deformation
P	load
P_{max}	maximum load
P_Y	critical load to induce 'yield' in blunt contacts
P_C	critical load to induce fracture in blunt contacts
R	radius of curvature
S_H	sensitivity to hydration
σ_Y	yield strength
T	fracture toughness
σ	strength
ν	Poisson's ratio
Υ	constraint factor
ξ_r	magnitude of residual stress field

A thorough understanding of how the arrangement and properties of individual components determine the performance of the 'bulk' materials is a prerequisite for any attempt at replicating their functionality. Hence, mechanical and structural characterisation methods traditionally developed for and applied to engineering materials are increasingly used to study biological materials. Among these, instrumented indentation has been particularly popular, due to the relative ease of use and its ability to accurately measure material properties on various length scales [6–8]. In biological materials, instrumented indentation is frequently used as a tool to relate structural changes, for example degree of mineralisation, to variations in material properties, such as fracture toughness, elastic modulus, and hardness. These properties, in turn, determine performance and hence the adaptive value of the structural changes in question, and therefore can shed light on critical structure–function relationships.

Fracture toughness, T, quantifies a material's resistance to crack initiation and propagation, and is superseding 'strength' as the property most associated with high-performance materials [9,10]. The hierarchical design and the ensuing complexity of biological materials necessitates a careful execution and application of the existing indentation methodology [11]. However, instrumented indentation remains an unrivalled method for the evaluation of toughness, in particular in biological materials which are frequently too small or irregular to allow the fabrication of 'standardised' specimen required for more conventional fracture tests [8].

The elastic modulus quantifies the resistance to elastic (reversible) deformation. In contrast to toughness, a large modulus is not necessarily desired (for example, adhesive structures need to be somewhat compliant), so that a direct link between its magnitude and performance is often not straightforward. Most studies assume isotropic behaviour of biological materials, i.e. a mechanical response that is independent of the testing direction (but see e.g. [12–15], where anisotropy in indentation was explicitly accounted for). However, it is frequently acknowledged that this assumption is more practical than accurate, and most of the literature hence reports an indentation modulus, E_I, which is equal to the plain strain modulus, E', if the modulus of the indenter is much larger than that of the material, and if the material is isotropic.

Hardness is probably the most commonly measured, but least understood of the material properties that can be assessed by instrumented indentation. Despite an increasing body of literature which has highlighted that hardness is a complex property that needs to be interpreted with care (see e.g. [6,16–20]), it is still often taken as a proxy for the resistance to 'plastic' (i.e. irreversible) deformation. This interpretation has been particularly popular in the more biological literature, perhaps because it provides a straightforward link to functional and hence ecological relevance. Hardness has long been a property of considerable importance in engineering materials: it is an index of strength [21] and – in combination with toughness – of brittleness [22]. Hardness is frequently associated with a material's resistance against being penetrated (i.e. elastically deformed), spread (i.e. irreversibly deformed) or scratched (i.e. fractured) [23]. Clearly, the physical mechanisms involved in these processes differ. What is the functional meaning of indentation hardness in biological materials?

In this article, this question is addressed by presenting a comprehensive meta-analysis of published data on indentation modulus, E_I, and indentation hardness, H_I, of diverse biological materials. We investigate the relationship between the two properties, which is then interpreted with a series elastic–plastic deformation model. This analysis provides guidelines for a careful interpretation and comparison of indentation hardness across and within biological materials, and highlights potential pitfalls. Next, we focus more specifically on the functional relevance of T, E_I and H_I for the avoidance and containment of mechanical damage introduced by 'blunt' or 'sharp' contacts,[1] and discuss the competition between quasi-plasticity and brittle fracture in biological materials. Quasi-plasticity emerges as the dominant damage mode for most biological materials, which is not solely controlled by indentation hardness, but by its ratio with the indentation modulus.

2. Analysis & discussion

2.1. The correlation between indentation hardness and modulus

Indentation hardness and modulus of various biological materials were assembled from the literature ([24–91], all data are avail-

---

[1] The distinction between sharp and blunt contacts is somewhat arbitrary – sharp indenters are those with a steep angle of the tangent at the point of contact. Irreversible deformation will necessarily precede cracking if the contact is sharp, while the deformation can remain dominantly elastic prior to fracture if the indenter is blunt. To a first approximation, spheres are blunt, and conical, Vickers, Berkovich and cube corner indenters are sharp.