Electronic Supplementary information

for

Can hyperelastic material parameters be uniquely determined from indentation experiment?

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Section I.

Buckingham Π theorem was adopted for the analysis. The theorem describes how an equation involving *k* physical variables can be rewritten into an equivalent equation of $k - m$ dimensionless Π functions, where *m* is the number of variables with independent dimensions. Because the procedures of dimensional analysis for different shapes of indenters are similar, we only present the analysis for conical indenter as a representative.

For conical indenter, *f* - *d* relationship can be expressed by the following function:

$$F\left(f_{\mathrm{C}}^{\mathrm{OG}}, d, G_0, \theta, \gamma_1, \ldots, \gamma_{2N-1}\right) = 0 \tag{S1}$$

The physical quantities in eqn (S1) constitute a variable vector:

$$\mathbf{q} = \left\{f_{\mathrm{C}}^{\mathrm{OG}}, d, G_0, \theta, \gamma_1, \ldots, \gamma_{2N-1}\right\}^{\mathrm{T}} \tag{S2}$$

Table S1. Dimensions of the variables in *f* – *d* relationship.

Variable	$[f]$	$[G]$	$[\gamma]$	$[d]$	$[a]$	$[\theta]$	$[r]$
Dimension	LMT^{-2}	$\mathrm{L}^{-1}\mathrm{MT}^{-2}$	none	L	L	none	L

Dimensions of the independent variables of **q** are listed in Table S1 and can be expressed through basic dimension vector **X** and dimension matrix **Ω** as shown in eqn (S3) and eqn (S4) below:

$$\left[q_j\right] = \prod_{i=1}^{3} X_i^{\Omega_{ij}}, \; j = 1, \ldots, 2N+3 \tag{S3}$$

where the bracket '$[\;]$' denotes the dimension of a physical quantity. Basic dimension vector **X** is expressed as $\{\mathrm{L,M,T}\}^{\mathrm{T}}$, where L, M and T represent the dimensions of length, mass and time, respectively. Dimension matrix $\mathbf{\Omega}$ takes the following form:

$$\mathbf{\Omega}=\begin{bmatrix} 1 & 1 & -1 & 0 & 0 & \cdots & 0 \\ 1 & 0 & 1 & 0 & 0 & \cdots & 0 \\ -2 & 0 & -2 & 0 & 0 & \cdots & 0 \end{bmatrix} \tag{S4}$$

where the j-th column corresponds to the dimensions of the corresponding independent j-th variable q_j. Since $\mathbf{\Omega}$ as shown in eqn (S4) is a matrix of rank 2, a natural choice of $(2N+3)-2=2N+1$ solutions to the linear equation $\mathbf{\Omega}\cdot\mathbf{y}=\mathbf{0}$ is shown in eqn (S5) below:

$$\begin{cases} \mathbf{y}_1=\{-1,2,1,0,\cdots,0\}^{\mathrm{T}} \\ \mathbf{y}_2=\{0,0,0,1,0,\cdots,0\}^{\mathrm{T}} \\ \mathbf{y}_3=\{0,0,0,0,1,0,\cdots,0\}^{\mathrm{T}} \\ \vdots \\ \mathbf{y}_{2N+1}=\{0,0,0,0,\cdots,1\}^{\mathrm{T}} \end{cases} \tag{S5}$$

Based on eqn (S5), the dimensionless functions $\Pi_i\ (i=1,\cdots,2N+1)$ are denoted as:

$$\begin{cases} \Pi_1=\gamma_1 \\ \vdots \\ \Pi_{2N-1}=\gamma_{2N-1} \\ \Pi_{2N}=\left(f_{\mathrm{C}}^{\mathrm{OG}}\right)^{-1}G_0d^2 \\ \Pi_{2N+1}=\theta \end{cases} \tag{S6}$$

Then eqn (S1) can be written equivalently as a function of Π_i.

$$\Pi_{2N}=\Phi\left(\Pi_1,\cdots,\Pi_{2N-1},\Pi_{2N+1}\right) \tag{S7}$$

Substituting eqn (S6) into eqn (S7), the $f-d$ relationship could be expressed by Π functions as:

$$f_{\mathrm{C}}^{\mathrm{OG}}=G_0d^2\Pi_{\mathrm{C}}^{\mathrm{OG}}\left(\gamma_1,\cdots,\gamma_{2N-1},\theta\right) \tag{S8}$$

where $\Pi_{\mathrm{C}}\left(\gamma_j,\theta\right)=\Phi^{-1}\left(\gamma_j,\theta\right)$.

For spherical and flat indenters, the expressions of $\mathbf{q}$ are expressed as

$\left\{f_{\mathrm{S}}^{\mathrm{OG}}, G_0, \gamma_1, \ldots, \gamma_{2N-1}, \sqrt{rd}d, d/r\right\}^{\mathrm{T}}$ and $\left\{f_{\mathrm{F}}^{\mathrm{OG}}, G_0, \gamma_1, \ldots, \gamma_{2N-1}, da, d/a\right\}^{\mathrm{T}}$, respectively. Repeating above procedure, the $f-d$ relationships expressed by Π functions could be obtained as shown in eqn (1).

Section II.

To ease reading, we adopt a two-parameter Arruda-Boyce model to illustrate the problem. The $\Psi-\Delta_l$ relationships of spherical and flat indenters are then reduced into the following form:

$$\Psi_l^{\mathrm{AB}} = G_0 \Pi_l^{\mathrm{AB}}\left(\gamma_1, \Delta_l\right),\ l = \mathrm{S} \text{ or } \mathrm{F} \tag{S9}$$

Applying the method of least squares (MLS) to fit eqn (S9) with *n* points of experimental data, *S*, the sum of squared residuals, is calculated as:

$$S = \sum_{i=1}^{n}\left[{}^{i}\Psi_l^{\mathrm{AB}} - G_0 \Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)\right]^2 \tag{S10}$$

where the left superscript *i* takes the value from 1 to *n*. To minimize *S*, following conditions shall be satisfied:

$$\begin{cases} \dfrac{\partial S}{\partial G_0} = \displaystyle\sum_{i=1}^{n} 2\left[{}^{i}\Psi_l^{\mathrm{AB}} - G_0 \Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)\right]\Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right) = 0 \\ \dfrac{\partial S}{\partial \gamma_1} = \displaystyle\sum_{i=1}^{n} 2\left[{}^{i}\Psi_l^{\mathrm{AB}} - G_0 \Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)\right] G_0 \dfrac{\partial \Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)}{\partial \gamma_1} = 0 \end{cases} \tag{S11}$$

If γ_1 and Δ_l are decoupled as $\Pi_l^{\mathrm{AB}}\left(\gamma_1, \Delta_l\right) = \Pi_l^{\mathrm{AB}\_\gamma_1}\left(\gamma_1\right)\Pi_l^{\mathrm{AB}\_\Delta_l}\left(\Delta_l\right)$, because $\Pi_l^{\mathrm{AB}\_\gamma_1}\left(\gamma_1\right)$ is not related to a particular experimental point (not left-superscripted by *i*) and then can be taken out of the summation, eqn (S11) can be rewrite as:

$$\begin{cases} \dfrac{\partial S}{\partial G_0} = 2\Pi_l^{\mathrm{AB}\_\gamma_1}(\gamma_1)\sum_{i=1}^{n}\left[{}^{i}\Psi_l^{\mathrm{AB}} - G_0\Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)\right]\Pi_l^{\mathrm{AB}\_{}^{i}\Delta_l}\left({}^{i}\Delta_l\right) = 0 \\ \dfrac{\partial S}{\partial \gamma_1} = 2G_0\dfrac{\partial \Pi_l^{\mathrm{AB}\_\gamma_1}(\gamma_1)}{\partial \gamma_1}\sum_{i=1}^{n}\left[{}^{i}\Psi_l^{\mathrm{AB}} - G_0\Pi_l^{\mathrm{AB}}\left(\gamma_1, {}^{i}\Delta_l\right)\right]\Pi_l^{\mathrm{AB}\_{}^{i}\Delta_l}\left({}^{i}\Delta_l\right) = 0 \end{cases} \quad \text{(S12)}$$

Obviously, the two equations in eqn (S12) are equivalent, which indicates infinite sets of $\{G_0, \gamma_1\}$ can satisfy eqn (S12). Therefore, $\{G_0, \gamma_1\}$ cannot be uniquely determined as long as γ_j and Δ_l are decoupled.

If γ_j and Δ_l are coupled in Π function, the two equations in eqn (S11) are linearly independent, which leads to a solution space of 0-dimension. Hence, unique solution could exist. However, the form of Π function could be complicated for hyperelastic indentation problem. If it is ill-posed, the solution can still be non-unique. Therefore, the determination of $\{G_0, \gamma_1\}$ can only be regarded conditionally unique.

The above analysis is based on MLS. For other types of fitting methods, the analysis procedures shall be similar.

Section III.

As shown in eqn (1), for single-parameter hyperelastic model, the $f-d$ relationship can be written as:

$$\begin{cases} f_{\mathrm{C}} = G_0 d^2 \Pi_{\mathrm{C}}\left(\rho_{\mathrm{C}}\right) \\ f_{\mathrm{S}} = G_0\sqrt{\rho_{\mathrm{S}} d}\, d\Pi_{\mathrm{S}}\left(\Delta_{\mathrm{S}}\right) \\ f_{\mathrm{F}} = G_0 \rho_{\mathrm{F}} d \Pi_{\mathrm{F}}\left(\Delta_{\mathrm{F}}\right) \end{cases} \quad \text{(S13)}$$

For a displacement-controlled experiment, the experimental error is in the force f. Therefore, the condition number of hyperelastic parameter G_0, which is written as $G_0 f'(G_0)/f(G_0)$ by definition, is solved as 1. This conclusion is independent of the form of

Π function and the shape of indenter.

Table S2. The posedness of hyperelastic indentation problems with different combinations of indenter shapes and hyperelastic models.

No. of parameter / Indenter Shape	Single-parameter	Two-parameter	Multiple-parameter
Conical	well	ill	ill
Spherical	well	For the conditionally unique problem, depends on the stability; otherwise ill.	
Flat	well		