Contact mechanics of soft granular materials with capillary interaction

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Abstract. Engineering applications of soft granular materials require accurate characterisation of their mechanical properties (*e.g.*, Young's modulus). However, the formation of liquid bridges between the high-water-content surface and substrate introduces capillary forces, which adds complexity to contact mechanics and potentially compromises measurement accuracy. In this study, we conducted indentation tests on individual hydrogel particles to investigate the effects of capillary force on force-displacement curves. These effects were analysed by quantifying and tracking the curvatures of the liquid bridge during tests from image processing. Our results demonstrate that Young's modulus estimation from raw data was overestimated compared to that obtained after accounting for capillary contributions. By isolating and correcting for capillary force contributions, this study provides an improved and reliable experimental method for accurately characterising the mechanical properties of soft granular materials, ensuring for safe and effective use in various critical applications.

1 Introduction

Soft granular materials, such as hydrogel spheres, have recently attracted significant attention due to their biocompatibility, biodegradability, self-healing capabilities, and high-water content. These characteristics make hydrogels particularly suitable for a range of applications, including biomedical devices, tissue engineering and soft robotics [1-2]. To ensure proper functionality, compatibility, reliability, and safety across these diverse applications, it is essential to accurately characterize their mechanical properties, such as Young's modulus. Additionally, surface characteristics and viscoelastic behaviour play critical roles in determining sensing accuracy and durability of robotic systems [3].

Among various techniques for characterizing the mechanical properties of soft materials, indentation testing has been widely adopted due to its precision and experimental simplicity [4]. This method involves measuring the applied load as a function of controlled displacement. Compared to other mechanical testing methods such as bulk compression, rheometry, and atomic force microscopy (AFM), indentation is particularly suitable for probing the mechanics of small, discrete soft granular systems [5].

Classical contact models, such as Hertz theory, are commonly employed to extract Young's modulus from force-displacement data [6]. The accuracy of these models relies on the critical assumption that all measured forces result solely from linear elastic deformation. However, in the case of highly hydrated soft particles like hydrogels, this assumption can break down due to capillary force arising from the forming liquid bridges at the contact interface. Capillary force becomes particularly prominent at small scales, where surface effects outweigh bulk mechanical behaviour [7], or for soft materials where the relative contributions from capillary interaction and elastic response are comparable. In addition to introducing attractive forces, liquid bridges induce localised deformation within the soft material, further complicating the interpretation of the measured mechanical response [8]. As a result, neglecting these capillary contributions can lead to substantial errors in the estimated material properties. Yet, the existing indentation tests could not isolate the capillary effects from elastic responses through the measured force-displacement curves.

Although considerable progress has been made in understanding capillary forces between soft planar surfaces [8], much less is known about capillary interactions between soft granular particles. This gap is particularly relevant as spherical hydrogels are increasingly employed in applications such as self-healing materials and injectable biomaterials [1]. In these systems, capillary interactions between individual particles can significantly alter aggregation behaviour and reconfigurability [9]. Therefore, it is crucial to understand how capillary forces influence the soft particle deformation during indentation testing and to distinguish the capillary contributions from purely mechanical responses.

This study aims to investigate the effect of capillary bridges on the mechanical characterisation of hydrogel particles through indentation tests. By capturing both the measured combined mechanical load and the capillary contribution derived from liquid bridge geometry, we quantitatively assess how liquid bridges affect the

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apparent stiffness measurements of soft particles. This work underscores the necessity of isolating capillary effects to enable accurate mechanical characterisation of soft granular materials.

2 Methodology

2.1 Materials

Two primary materials were used in this study: crosslinked polyacrylamide hydrogel spheres as the testing materials and polydimethylsiloxane (PDMS) as the flat substrate. In the present work, the PDMS layer acts as the supporting substrate for indentation tests of hydrogel particles. Due to its significantly higher stiffness, it can be considered mechanically rigid compared to the hydrogel particles. The dry diameter of the hydrogel particles ranges from 2 to 2.5 mm. After being immersed in demineralised water for 24 hours in a sealed container to prevent contamination, the fully swollen spheres reached diameters in the range of 15-18 mm. The swelling process was carried out in a sealed container to prevent contamination. The PDMS substrate was prepared using Sylgard 184 (Dow Corning) by mixing the base agent and curing agent at a 10:1 weight ratio. The mixture was manually stirred with a glass rod for 10 minutes for uniform dispersion and then degassed in a vacuum chamber for 15 minutes to remove air bubbles. It was subsequently cast into a custom quartz-glass mould (30 mm × 30 mm × 5 mm) and cured at 23 °C for 24 hours.

2.2 Experimental setup

A custom-developed loading system was employed to monitor the response of hydrogel particles during indentation experiments, as shown in Fig. 1. The system includes an electronic scale (A&D, HR-250 AZ, with a resolution of 0.0001g), an industrial digital camera (DAHENG, MER2-2000-19U3M, with a spatial resolution of 2.4 μm / pixel), a linear translation stage (Thorlabs, MTS25/M-Z8), an optical platform (flatness: ±0.05 mm over 0.36 m^2), a white LED light (JSIONX, JS-DBL209-318).

A 3D-printed board was mounted onto the translation stage to provide a rigid horizontal platform, and a flat glass plate was affixed to the underside of this board and served as the top indenter in the compression experiment. A custom-designed 3D-printed holder was tailored to the size of hydrogel to prevent lateral movement and ensure consistent alignment. A transparent ruler was affixed vertically to the side of the setup to serve as a scale reference in the captured images.

2.3 Test procedures

A total of three individual hydrogel particles were tested, each under identical loading conditions. The PDMS substrate was placed on the electronic scale, and illumination sources were activated. Prior to each experiment, any residual water was carefully removed from the PDMS surface, and the electronic scale was re-zeroed. The diameter of each hydrogel particle was measured and gently placed onto the PDMS substrate.

The indentation experiment was controlled using a Python script that simultaneously triggered data acquisition from the scale, translation stage and camera. During the indentation process, the glass plate attached to the 3D printed board was lowered at a constant rate of 0.01 mm/s to compress the hydrogel by 0.75-0.9 mm (approximately 5% of its initial diameter). Once the target displacement was achieved, the platform was immediately unloaded at a speed of 0.1 mm/s.

The scale and the translation stage were used to record weight and displacement data at 100 ms intervals. The industrial camera equipped with a 25 mm lens was aligned horizontally with the height of the capillary bridge between hydrogel and PDMS and captured images at 5-second intervals. The camera, hydrogel centre, and LED were aligned along a horizontal axis, and the laser, hydrogel, and optical platform shared the same horizontal plane to ensure accurate visualization of the liquid bridge profile.

2.4 mage and data processing

The recorded weight and height data were used to calculate scale force (F_{scale}) and displacement (δ). To analyse the geometry of the capillary bridge, side-view images were converted to grayscale, then binarized and

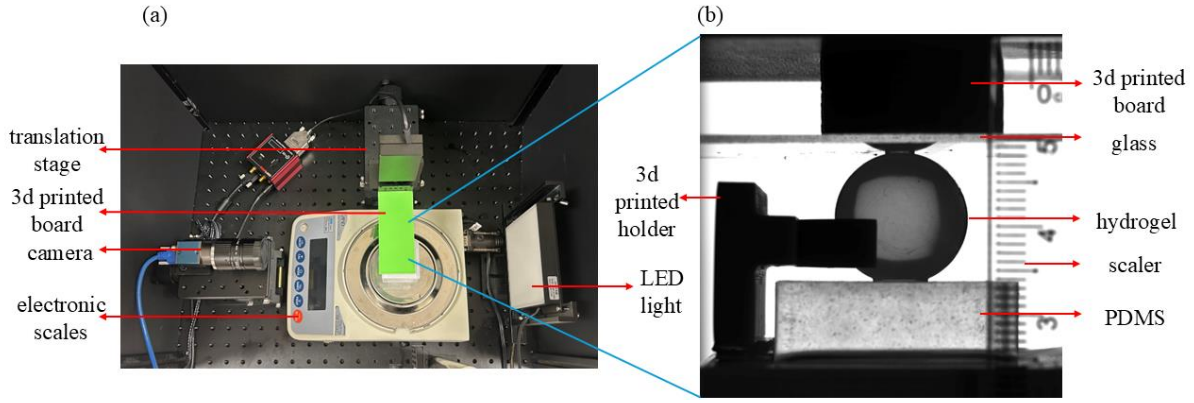

Fig. 1. (a) Overview of the custom-developed test setup (top view); (b) A typical experimental image captured by the camera.

denoised using Gaussian blur and morphological operations. Edge detection was then performed using the Canny method to extract the liquid-air interface. Each of these time-series images, captured at 5-second intervals, was processed to extract the liquid-air interface and fitted with circular arcs using a least-squares method to determine the principal radii at each time step. The first principal radius (R_1) was determined as the average of the fitted curvatures on both left and right sides of the liquid bridge. The second principal radius (R_2) was defined as the horizontal distance between the two fitted curves, representing the span of the liquid bridge, as illustrated in Fig. 2. All radii were converted from pixel units to millimetres using the scale factor determined from Fig. 1.

3 Results

The contact force (F_{cont}) acting on the hydrogel during indentation was calculated as the sum of the scale-measured force (F_{scale}) and the capillary force (F_{cap}):

$$F_{cont} = F_{scale} + F_{cap}, \tag{1}$$

$$F_{cap} = \pi \cdot {R_2}^2 \cdot \Delta p + 2\pi R_2 \cdot \gamma, \tag{2}$$

where $\Delta p = \gamma \left(\frac{1}{R_1} + \frac{1}{R_2}\right)$ is the pressure difference across the interface. Both R_1 and R_2 were obtained from a time series of images taken every 5 seconds, thus Δp varies with the evolving liquid bridge geometry and contact area. The surface tension γ is taken as constant (72.8 mN/m) under controlled lab conditions of 40-60% humidity and 23 °C.

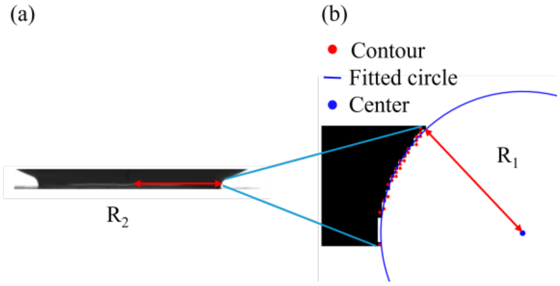

Fig. 2. Image processing to extract principal radii based on cropped liquid bridge (a) R_2; (b) R_1.

The force-displacement responses derived from both F_{scale} and F_{cont} are presented in Fig. 3. The contact force consistently exceeds the scale-measured force at the same indentation depth, indicating that the capillary force contributes significantly to the overall force response. This discrepancy highlights the potential for overestimating material stiffness when capillary effects are neglected. The variation in capillary forces with displacement were also reported in Fig. 3, represented as the gap between these two force curves. The trend is relatively constant due to the limited range of displacement, as our current focus is in the linear elastic region.

To quantify the effect of capillary on the measured stiffness, both F_{scale} and F_{cont} were fitted using Hertz contact theory:

$$F = k \cdot (\delta - \delta_0)^{\frac{3}{2}}, \tag{3}$$

$$k = \frac{4}{3} E R^{1/2}, \tag{4}$$

where k is the fitting parameter representing stiffness, δ is half of the displacement from top indenter during compression, measured relative to the initial position of the hydrogel particle, and R is 5% of the diameter of each hydrogel. In Eq. (3), δ_0 is the threshold displacement that marks the onset of mechanical contact. Both k and δ_0 were obtained through curve fitting and were independently determined for F_{cont} and F_{scale}. The value of δ_0 reflects the true elastic deformation contact point under different forces, while the variation k highlights the influence of capillary interactions on material stiffness.

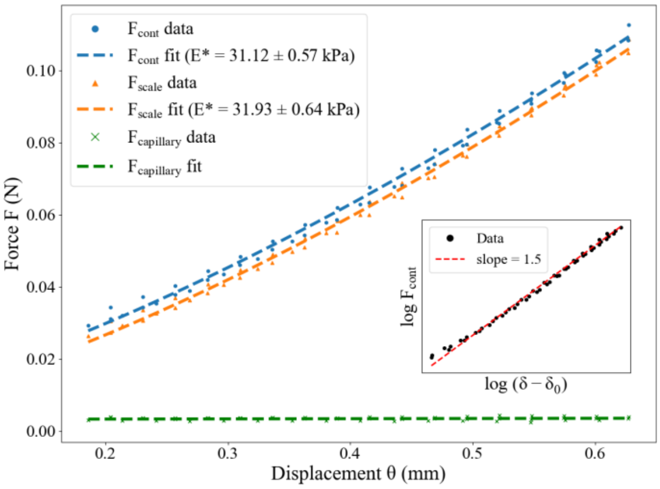

Fig. 3. Representative force–displacement curves for three hydrogel particles, fitting with Hertz theory. The inset shows a log–log plot of the corrected contact force versus displacement.

The fitting results from three hydrogel particles yielded an apparent modulus of E_{scale} = 31.93 ± 0.64 kPa based on scale forces, and an intrinsic modulus of E_{cont} = 31.12 ± 0.57 kPa based on capillary-corrected contact forces. The apparent stiffness was consistently higher than the intrinsic value, demonstrating that neglecting capillary contributions leads to a systematic overestimation of material stiffness.

Experimental comparison is crucial given the small magnitude and sensitivity of capillary forces to system scaling. However, most prior studies have examined either rigid particles at the microscale or stiff spheres on soft planar supports where deformation primarily occurs in the substrate [10-11]. In contrast, our configuration involves millimetre-scale soft spheres on a rigid substrate, shifting the deformation to the particle side and introducing distinct capillary-elastic coupling that is fundamentally different and not directly comparable to those earlier configurations. Still, our measured capillary forces (2-5 mN) agree closely with benchmarks from a similar soft material system: Butt *et al.* reported effective moduli below 50 kPa in this capillary force range [12]. This close agreement indicates that our measurements are reasonable for the present system.

4 Discussions

Although our experimental approach reliably characterises the mechanical behaviour of soft granular materials, future work should incorporate additional variables to derive more generalised property estimates. In particular, we tested only commercial hydrogel spheres on a single substrate, leaving both mechanical and interfacial properties fixed. Yet substrate stiffness, surface wettability, and hydrogel properties (*e.g.*, crosslinking ratio, water content) critically affect the force response and contact mechanics. Variations in these properties can alter liquid bridge geometry, deformation magnitude, and thus the capillary contributions. Future studies should therefore systematically vary substrate modulus and surface wettability, and compare hydrogels with different swelling ratios, network structures, and viscoelastic characteristics.

Additionally, this study serves as an initial step toward highlighting the role of capillary forces in soft particle contacts. To remain within the small-deformation regime where Hertz contact theory is applicable, the indentation depth was restricted to 0.75-0.9 mm (~5% hydrogel diameter). This approach enables the use of Hertzian predictions as a mechanical baseline, allowing the capillary contribution to be inferred by comparison with experimental measurements. A more comprehensive framework could be developed by incorporating the Johnson–Kendall-Roberts (JKR) theory, which treats capillary attraction as an effective adhesion energy. By coupling JKR-like adhesion with evolving Laplace pressure, future models would be capable of capturing both elastic deformation and interfacial dynamics in soft capillary contacts.

5 Conclusion

This study investigated the influence of capillary forces on the mechanical characterisation of granular soft materials using indentation tests. The presence of liquid bridges introduces additional attractive forces, which altered the measured force-displacement response. As a result, Young's modulus estimation from raw data was significantly overestimated compared to that obtained after counting for capillary contributions. These findings highlight the necessity of separating capillary and mechanical contributions when analysing the mechanical properties of soft granular materials.

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