



## Review

## Innovative evolution of buckling structures for flexible electronics

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## ABSTRACT

A buckling structure is a crucial element for materials used in wearable electronics by taking a topographical approach to solve a spatial constraint. This review introduces various buckling structures, with the corresponding theory and fabrication, for the latest materials or devices, including secondary batteries, supercapacitors, sensors, organic thin film transistors, polymer light-emitting diodes, and organic light emitting diodes, all of which have attracted much attention recently in journals. This review demonstrates how various buckling structures can be practically integrated into functional materials to enlarge the active area, to enhance flexibility or stretchability, and to improve unique functionality itself without loss.

## 1. Introduction

A buckling structure is defined as a topological pattern resulting from a mathematical or physical instability and is typically used to overcome spatial constraints for various flexible devices while maintaining their original functionalities [1–5]. Such topological properties, generated by micro/nano wrinkling, folding, creasing, and delaminating, can be exploited for use in smart microfluidic designs, antifouling, and specific functional surfaces [6–11]. Therefore, various buckling structures are frequently implanted to dynamically support stretchability, tunability, and multiple functionality for the fabrication of flexible devices or composite materials such as sensors and robotic skin [12–15]. Even though highly advanced functional materials have been developed, their brittle properties prevent the use of such materials from stretchable or wearable electronics. Combination of the rigid functional materials with stretchable substrates in the form of successful buckling structure enables the rigid functional materials to be deformation resistive as if they were rubber-like elastomers. [16–18]. Indeed, buckling materials potentially available for new functional composites or stretchable electronics have been continuously introduced [19–21]. To maintain the original functionality of the surface layer when stretched to extremes, representative buckling structures such as miura pattern, serpentine, and notch-islands have been developed for enhanced flexibility, stretchability, and stability of devices or materials with minimal functional sacrifice [22–24].

Unstable and indurated surface structures by high compression forces are usually generated through the prestrain/release, axial

compression, and various chemical reactions between soft substrates and relatively rigid surface materials [25–28]. Periodic structural deformation due to the load and direction of the mechanical distortion of elastomeric soft substrates, including polydimethylsiloxane (PDMS), polypyrrole (PPy), and polyethylene terephthalate (PET), occur in a divergent manner [29–31]. From a more practical point of view, preparation methods for functional surface materials on the substrates, such as stretching-coating-releasing process, spin coating, lithography, ultraviolet/ozone (UV/O<sub>3</sub>) radiation, osmotic swelling, sputtering, plasma, and their combinations in many cases, strongly affect the featuring properties of a buckling structure [1,6,25,32–36].

This paper conducts a brief theoretical review of buckling formation with introducing buckling wave parameters and basic buckling patterns, classification of buckling materials in terms of organic or inorganic materials, fabrication methods of the composite buckles to minimize spatial constraint with enhanced adhesion strength between the substrate and the surface layer, and finally a review of applications for the buckles in flexible display such as organic thin-film-transistor (OTFT), organic light emitting diode (OLED), and polymer light-emitting diode (PLED), and stretchable energy harvesting/storage devices including supercapacitors, secondary batteries, smart windows, and organic photovoltaic cells (OPVs) [37–43].

## 2. Basic theoretical background and fundamental patterns of buckling structure

A buckling structure results from a change in topological structure

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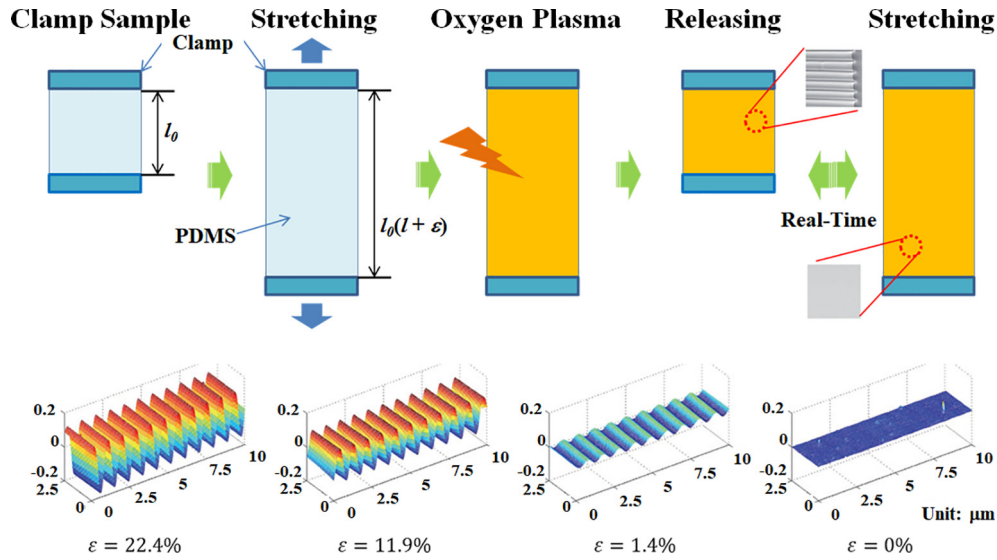


Fig. 1. A sketch showing preparation of the mechanically tunable buckling structure using PDMS substrate at various  $\varepsilon$  strains. The actual preparation process consecutively follows stretching, O<sub>2</sub> plasma treating, and releasing the PDMS substrate. Adapted from [47].

commonly dependent on the magnitude of an applied compressive stress [44,45]. For example, an ordered, periodic, one-dimensional wavy pattern is formed in a yarn-type buckling composite, and the buckling wavelength ( $\lambda_b$ ) and amplitude ( $A_b$ ) of the pattern are important parameters determining the buckling efficiency. A short  $\lambda_b$  with a large  $A_b$  means a high buckling efficiency. If such buckling structure were employed to prepare an energy storage material, the buckling efficiency directly determines the capability of the energy storage of the yarn and effectiveness of the compressive or tensile stress-induced energy dissipation by the buckling motion [32,46]. Usually, buckles occur along the principal axis as the buckling material is stretched and subsequently released, i.e., a stripe orientation of the buckling structure can be formed as the material is contracted perpendicularly to the direction of the elongation by a compressive stress. Shape and size of the buckling structure can be numerically expressed by determining two major buckling wave parameters ( $\lambda_b$  and  $A_b$ ). These parameters strongly depend on an externally applied axial stress. Examples include that the buckling parameters of the most frequently-used PDMS-based stretchable substrate in an oxygen plasma environment vary with the applied strain ( $\varepsilon$ ) (Fig. 1) [47–49].

The  $\lambda_b$  and  $A_b$  of the sinusoidal buckling structure in a two-component composite (a soft substrate and a functional surface layer) are given by Eqs. (1) and (2), respectively:

$$\lambda_b(E_s, E_f, h_f) = 2\pi h_f \left( \frac{\bar{E}_f}{3E_s} \right)^{1/3} \quad (1)$$

$$A_b(E_s, E_f, h_f, \varepsilon) = h_f \left( \frac{\varepsilon}{\varepsilon_c} - 1 \right) \quad (2)$$

where  $h$  is the thickness,  $\bar{E} = E(1 - \nu^2)$  is the planar modulus,  $E$  is Young's modulus,  $\nu$  is Poisson's ratio,  $\varepsilon$  is the strain by the compression force, and subscripts  $f$  and  $s$  represent the surface layer and the soft substrate, respectively [46]. Note that the critical strain of the buckle is  $\varepsilon_c = -0.25(3E_s/E_f)^{2/3}$ . In result, both  $\lambda_b$  and  $A_b$  are functions of  $E_s$  and  $E_f$ , and are linearly proportional to  $h_f$ .  $\varepsilon$  only affects  $A_b$ .

Other factors affecting the buckles' characteristics except the two major parameters may include  $E$ ,  $h$ , and the interfacial properties between the substrates and functional surface layers. Conventionally, a buckle composed of a viscoelastic polymer substrate and a thin surface is transformed by biaxial compression in a condition in which the critical moduli  $E_c (= E_f/E_s)$  is less than  $10^2$ , and the resulting multi-

periodic morphology is retained until an  $E_c$  of around  $10^3$ . Finally, the buckles are released at  $E_c = 10^4$ . In the buckles that are isotropically formed by the biaxial strain, the  $\lambda_b$  of the composite material depends on  $h_f$  and eventually on the mechanical resistance of the surface layer. Hence, the buckling structure-specific morphology will fail if the applied biaxial strain is larger than a critical point. The thicker the surface layer, the less resistive to structural deformation [50]. Practically, it is possible to measure the critical value of the maximum compressive stress  $\sigma_c$  (Pa) and the  $\lambda_b$  (m) of the associated buckle with assuming that the buckles are aligned perpendicular to the direction of the compressive stress and no serious heat effect during the surface layer deposition on the soft substrate is observed, as given by Eqs. (3) and (4), respectively:

$$\sigma_c \approx 0.52 \left( \frac{E_f}{1-\nu_f^2} \right)^{1/3} \left( \frac{E_s}{1-\nu_s^2} \right)^{2/3} \quad (3)$$

$$\lambda_b \approx 4.36 h_f \left( \frac{E_f(1-\nu_s^2)}{E_s(1-\nu_f^2)} \right)^{1/3} \approx 4.4 h_f \left( \frac{E_f}{E_s} \right)^{1/3} \quad (4)$$

where  $h_f$  represents the thickness of the surface layer [51].

The compressive stress  $\sigma$  can be applied to the buckling material from biaxial directions at the same time or sequentially. While a heterogeneous buckling pattern is obtained by the simultaneous biaxial compressive stress, a homogeneous pattern is obtained by the consecutive application of the stress from one direction to another. In addition, a hierarchical buckling structure can be isotropically structured depending on the buckling density and the directional distribution of the buckles (Fig. 2) [52].

### 3. Categories of the buckling materials

Buckles can be constructed from a variety of different material combinations and they can be categorized into three major groups: group A-organic surface layer/inorganic substrate; group B-organic surface layer/organic substrate; group C-inorganic surface layer/organic substrate. Thermal expansion or contraction of a rigid inorganic substrate causes tensile or compression forces to the elastomeric surface layer. The thermally-actuated graphene sheet on the SiO<sub>2</sub>/Si substrate creates of one- or two-dimensional wavy patterns depending on the opening's shape of the substrate and is useful to comprehend the working mechanism of the graphene-based electronics (group A

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