



Article

Evolution of Thin-Film Wrinkle Patterns on a Soft Substrate: Direct Simulations and the Effects of the Deformation History

Siavash Nikravesh  and Yu-Lin Shen * 

Department of Mechanical Engineering, University of New Mexico, Albuquerque, NM 87131, USA

* Correspondence: shenyl@unm.edu

Abstract: Surface wrinkling instability in thin films attached to a compliant substrate is a well-recognized form of deformation under mechanical loading. The influence of the loading history on the formation of instability patterns has not been studied. In this work, the effects of the deformation history involving different loading sequences were investigated via comprehensive large-scale finite element simulations. We employed a recently developed embedded imperfection technique which is capable of direct numerical predictions of the surface instability patterns and eliminates the need for re-defining the imperfection after each analysis step. Attention was devoted to both uniaxial compression and biaxial compression. We show that, after the formation of wrinkles, the surface patterns could still be eliminated upon complete unloading of the elastic film–substrate structure. The loading path, however, played an important role in the temporal development of wrinkle configurations. With the same final biaxial state, different deformation histories could lead to different surface patterns. The finding brings about possibilities for creating variants of wrinkle morphologies controlled by the actual deformation path. This study also offers a mechanistic rationale for prior experimental observations.

Keywords: thin film; wrinkle; deformation; surface instability; finite element analysis



Citation: Nikravesh, S.; Shen, Y.-L.

Evolution of Thin-Film Wrinkle Patterns on a Soft Substrate: Direct Simulations and the Effects of the Deformation History. *Nanomaterials* **2022**, *12*, 3505. <https://doi.org/10.3390/nano12193505>

Academic Editor: Jürgen Eckert

Received: 1 September 2022

Accepted: 5 October 2022

Published: 7 October 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

The formation of wrinkles (periodic buckles) on a thin film attached to a compliant substrate has received considerable attention in recent years. This form of surface instability is widely observed in nature and now increasingly encountered in flexible electronics and other functional devices [1–6]. Thin-film buckling can be triggered by the direct application of an in-plane compressive strain. It may also occur in response to applied tension, where a mismatch in lateral contraction between the thin film and a thick substrate forces the films into compression [7]. Wrinkling instability relaxes the compressive strain in the film layer, thus reducing the elastic strain energy. While the formation of wrinkles is traditionally viewed as an undesirable feature, it is increasingly exploited to enhance the mechanical and/or functional performances of energy and biomedical devices [8–13].

Theoretical solutions for the critical stress of wrinkle initiation, along with the wavelength and amplitude of the waveform, are available for the simplest forms of wrinkles [14–18]. The prediction of complex surface patterns resulting from the various loading, geometrical, and material conditions, however, will rely on computational means. Most numerical studies utilized the multi-step approach, with a pre-buckling step followed by a post-buckling analysis which essentially builds a waveform into the model [19–25]. The implementation can be laborious and lacks a true predictive capability. Other numerical techniques based on an applied fictitious/dummy load have been proposed [24,26–28], but multi-step analyses were still involved. Some of the recent studies followed similar approaches but focused more on wrinkling on curved composites [29,30]. Dynamic surface-instability simulations have also been conducted [31,32]; however, the verification and validation of the numerical outcomes are challenging, taking into account the lack of

reliable experimental results or closed-form solutions to loading-rate-dependent deformation instability problems. Recently, a peridynamics analysis of wrinkling instabilities has also been reported [33,34], but the validity of non-local theories (such as peridynamics) in reproducing the actual physical phenomenon is still uncertain. Moreover, defining some of the free parameters such as the “horizon size” is also cumbersome.

The analytical/numerical studies of surface instability were mostly performed under the consideration of an instantaneous and non-sequential in-plane uniaxial or biaxial loading. The potential effects of different loading sequences on instability patterns, though leading to an identical final state of strains, have not been investigated. It is worthy of mention that the possibility of such effects on wrinkling configurations have been implied in some earlier experimental studies [15,35].

We recently developed a practical finite element modelling approach to simulate wrinkle formation using the embedded imperfection technique [36,37]. The temporal wrinkling evolution can be captured in a straightforward manner, and the methodology has been employed to study complex surface patterns in the three-dimensional (3D) setting [38,39]. The present work extends our previous 3D analysis to cover the effects of the deformation history. Using the model system of a polymeric thin film on an elastomeric substrate under in-plane uniaxial and equi-biaxial compressions, we first studied the evolution of the surface instability patterns and how they might gradually disappear when the applied compression was reverted back to zero. Note that our modelling approach allows such process to be carried out in one simulation run. In the case of biaxial loading, we further investigated the effect of the deformation history by comparing the surface patterns resulting from simultaneous loading and sequential loading along the two axes. Our results revealed the sensitivity of the wrinkling patterns to the loading sequence and identified new forms of surface pattern evolution affected by the deformation history.

2. Numerical Model Description

The model consisted of a thin film bonded to a compliant substrate with an embedded imperfection. The film surface was square-shaped, and the substrate was sufficiently thick in comparison with the film. Figure 1 shows the problem geometry, boundary conditions, and directions of the applied displacement employed in the finite element analysis. All materials were chosen as linear elastic isotropic solids in the present study. The initial thickness of the film layer, t_f , was 0.1 μm . One of the common polymeric film materials used in organic optoelectronics and photovoltaics, namely, P3HT:PCBM [8], was considered. The Young's modulus of the thin film was $E_f = 7300 \text{ MPa}$ [8], and the Poisson's ratio was $\nu_f = 0.35$. The elastomeric substrate layer was PDMS, with Young's modulus $E_s = 2.97 \text{ MPa}$ [40] and Poisson's ratio $\nu_s = 0.495$ (set slightly smaller than 0.5 to mitigate potential convergence issues). As schematically shown in Figure 1, the problem domain contained a single embedded imperfection. It was a regular finite element at the exact center of the top layer elements in the substrate, immediately below the film–substrate interface. The embedded imperfection, while underneath the interface in the substrate domain, was assigned the material properties of the film layer. This approach has been systematically tested and proven to be a reliable way to trigger deformation instabilities in our earlier studies [38,39,41].

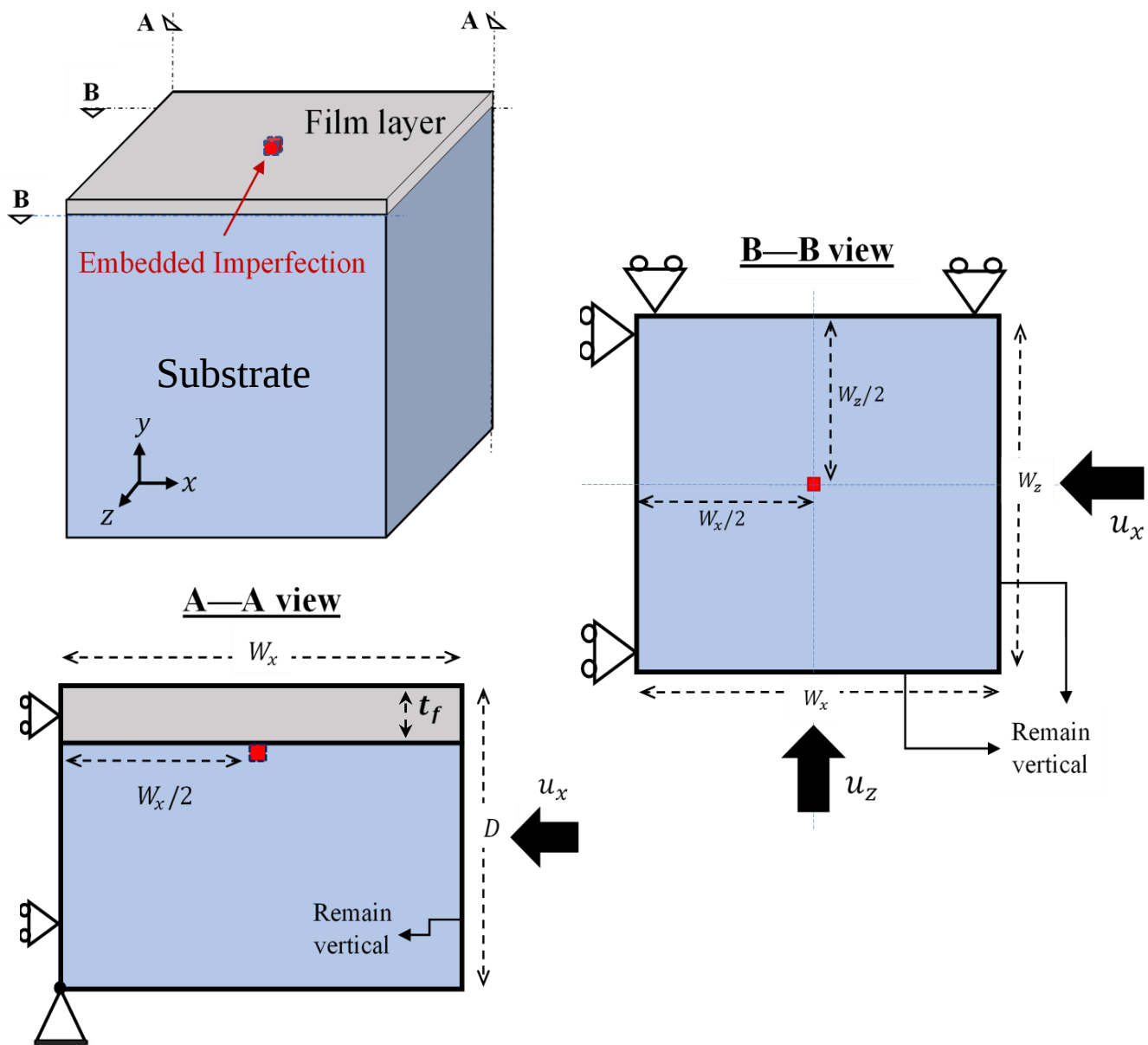


Figure 1. Schematics of the problem geometry, boundary conditions, and directions of the applied displacement [39].

Numerical simulations were performed using the finite element software ABAQUS (Version 2017, Dassault Systems Simulia Corp., Johnston, RI, USA). Throughout the model, 20-noded brick elements were used, with a uniform element distribution in the film layer (four layers of elements along the film thickness). An adapted element distribution existed in the substrate, with the element size increasing gradually from top to bottom. The in-plane mesh density of 10 elements per analytical sinusoidal wavelength was chosen to ensure mesh-convergent numerical solutions [38]. In addition to the mesh considerations, the displacement increment in the loading step was kept sufficiently small to avoid the potential increment dependency of the solutions [37]. The analyses were conducted via high-performance computing using the Message Passing Interface (MPI) parallelization technique. Preliminary calculations were carried out to ensure that the numerical results were independent of the parallel analysis procedures. Mesh convergence and model verification were also performed [38,39].

As shown in Figure 1, the roller boundary condition was imposed on the faces $z = 0$ and $x = 0$, and a corner point at the bottom of the substrate was fully fixed. The top face of the film and the bottom face of the substrate were kept traction-free. In addition, the faces $z = W_z$ and $x = W_x$ were constrained to remain vertical during the deformation. Therefore, the problem domain represented a unit cell of a large periodic structure [42]. The problem domain size was scaled by the value of the theoretical critical wavelength of the sinusoidal mode [14,16],

$$\lambda_{cr} = 2\pi t_f \left[\frac{(1 - \nu_s^2) E_f}{3(1 - \nu_f^2) E_s} \right]^{1/3} \quad (1)$$

The domain dimensions were $W_x = W_z = 10\lambda_{cr}$ ($=55.92 \mu\text{m}$) with the overall depth of $D = 5\lambda_{cr}$. The dimensions of $W_x = W_z = 10\lambda_{cr}$ were rationally chosen so that $10\lambda_{cr} \cong 7(\lambda_{cr})_{Cb}$, where $(\lambda_{cr})_{Cb} = \sqrt{2}\lambda_{cr}$ is the analytical wavelength of the square checkerboard instability mode under equi-biaxial compression [16]. Thus, the model size was well-suited to simulate various surface patterns in conformity with available analytical solutions. Displacement-controlled uniaxial and equi-biaxial compressions were considered. For uniaxial compression, a uniform displacement was applied in the x -direction only; for equi-biaxial compression, uniform displacements with equal magnitudes were applied concurrently in both the x - and the z -directions. The biaxiality ratio, BR, is defined as the ratio of the prescribed final z -displacement and x -displacement, in a given loading step. As a consequence, the uniaxial compression and equi-biaxial compression can be represented by $BR = 0$ and 1 , respectively. In this study, we also considered non-equi-biaxial loading with a BR value between 0 and 1.

The critical stress at the onset of the primary bifurcation, σ_{cr} , was analytically derived as [14,16,43]

$$\sigma_{cr} = \left[\frac{E_f}{4(1 - \nu_f^2)} \right] \left[\frac{3(1 - \nu_f^2) E_s}{(1 - \nu_s^2) E_f} \right]^{2/3} \quad (2)$$

assuming that the stress state was uniform (in the sufficiently thin film with the cross-section area of $t_f \cdot w_z$ (or $t_f \cdot w_x$), as shown in Figure 1). It has been postulated that the critical wrinkling stress in Equation (2) is applicable to any possible biaxial wrinkling mode (including the 1D sinusoidal form and the square checkerboard) [13]. The critical strain for the primary instability modes under pure uniaxial ($BR = 0$) and equi-biaxial compression ($BR = 1$) were reported as [14,16,23,44]

$$(e_{cr})_{1D} = \left(\frac{1}{4} \right) \left[\frac{3(1 - \nu_f^2) E_s}{(1 - \nu_s^2) E_f} \right]^{2/3} \quad (3)$$

and

$$(e_{cr})_{Cb} = \left[\frac{1}{4(1 + \nu_f)} \right] \left[\frac{3(1 - \nu_f^2) E_s}{(1 - \nu_s^2) E_f} \right]^{2/3} \quad (4)$$

where $(e_{cr})_{1D}$ and $(e_{cr})_{Cb}$ are the critical wrinkling strains of sinusoidal and square checkerboard wrinkles, respectively. It should be noted that Equations (3) and (4) were apparently obtained by dividing the critical stress in Equation (2) by, respectively, the plane strain modulus and the biaxial modulus of the film layer, namely, $(e_{cr})_{1D} = \sigma_{cr} / [E_f / (1 - \nu_f^2)]$ and $(e_{cr})_{Cb} = \sigma_{cr} / [E_f / (1 - \nu_f)]$. As can be recognized from Equations (3) and (4), considering the value of ν_f for the polymeric film material studied in this paper, $(e_{cr})_{1D} > (e_{cr})_{Cb}$. Moreover, for other cases of non-equi-biaxial compression, $0 < BR < 1$, closed-form theoretical solutions for the critical strain do not exist; however, from our earlier numerical studies [39], the critical strain associated with any BR lies within the range

between $(e_{cr})_{1D}$ and $(e_{cr})_{Cb}$ and varies with BR monotonically, with $(e_{cr})_{1D}$ being the upper limit at $BR = 0$, and $(e_{cr})_{Cb}$ being the lower limit at $BR = 1$. It was also demonstrated that the numerically obtained e_{cr} at any BR is slightly higher than the corresponding analytical values (if available) [39]. When presenting the results in the following sections, we used the numerically obtained critical strain in the x -direction at the onset of bifurcation, i.e., e_{cr} , for the corresponding BR loading state.

3. Results

3.1. Uniaxial Compression

We first considered a uniaxial compressive loading along the x -direction ($BR = 0$). Figure 2 shows the evolution of sinusoidal wrinkles during the loading and unloading phases. The color contours represent the extents of displacement in the out-of-plane (y) direction. In the loading phase (Figure 2a–c), the model was subjected to a compressive displacement from 0 to $-0.42 \mu\text{m}$ (corresponding to a compressive strain of -0.0075). The extent of the applied compression in terms of normalized strain was $e_{xx}/e_{cr} \cong 2.0$, where e_{xx} is the applied compressive strain in the x direction, and e_{cr} is the critical wrinkling strain obtained from the simulations [36]. As can be seen, fully developed sinusoidal wrinkles were obtained at the end of the loading phase (Figure 2c). Subsequently, the analysis was continued with an unloading phase, in that the applied strain was reverted back to zero (Figure 2c–e). The wrinkles tended to disappear gradually during unloading, and a fully flat surface was in place when the compressive strain was completely removed. The overall load–displacement response resulting from the simulation is shown in Figure 2f, with the model snapshots at different stages (Figure 2a–e) labeled along the curves. It is evident that the loading and unloading responses follow the same path. At the onset of instability during loading (and at the reversal of instability during unloading), there is a distinct change in slope of the load–displacement behavior.

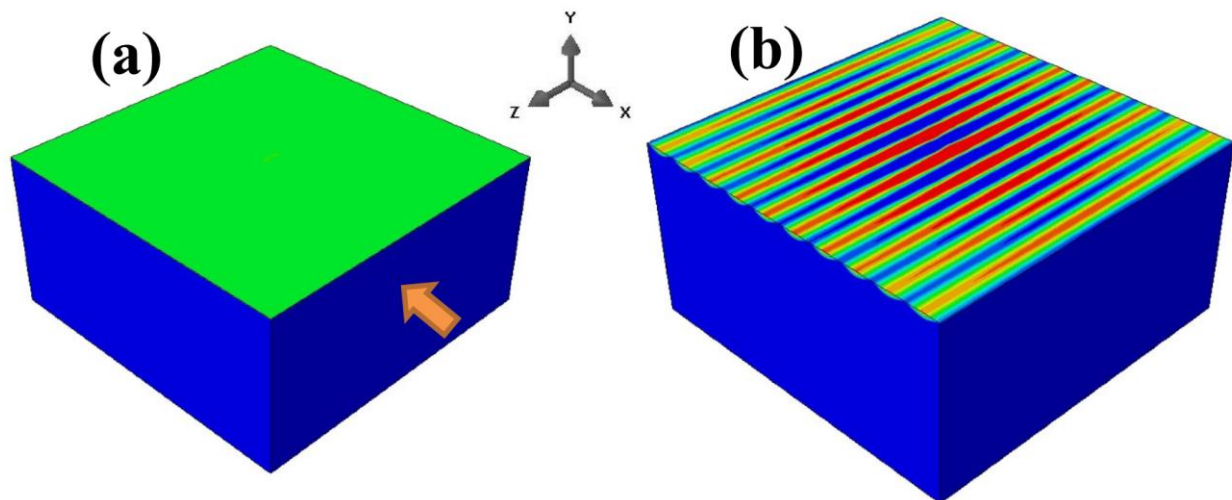


Figure 2. Cont.

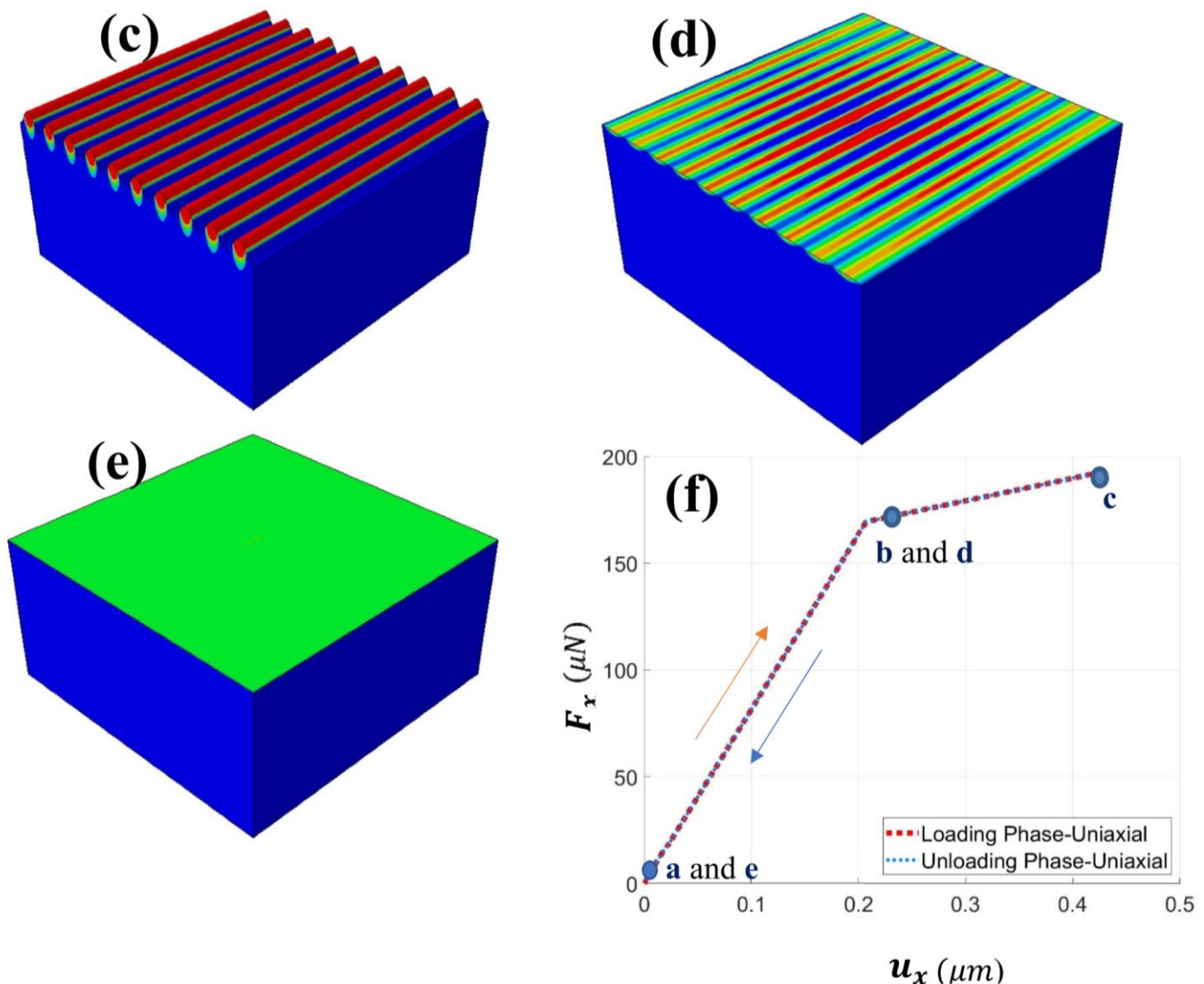


Figure 2. Snapshots of simulated surface patterns during the loading phase (from (a–c)) and unloading phase (from (c–e)) under uniaxial compression. (f) Overall load–displacement response for the entire deformation history, with the five stages (a–e) labeled along the curves.

3.2. Equi-Biaxial Compression

Attention was then turned to in-plane equi-biaxial loading along the x - and z -directions ($BR = 1$). The dominant mode of surface wrinkling at the onset of instability caused by an equi-biaxial loading is the square checkerboard pattern [15,39]. Although the pattern will eventually evolve into the labyrinth mode in the post-instability regime [39], the current section is limited to the primary mode of instability. Figure 3 shows the simulated evolution of the square-checkerboard wrinkles during loading (a–c) and unloading (c–e). The maximum equi-biaxial displacement of $-0.138 \mu\text{m}$ was applied during loading, which corresponded to $e_{xx}/e_{cr} \cong 1.03$, with e_{cr} being 0.0024. The development of a wrinkling pattern from the pre-instability flat face to the square checkerboard pattern can be clearly seen in Figure 3a–c. During the unloading phase (Figure 3c–e), the surface pattern gradually faded away, and the flat surface reappeared. Figure 3f shows the overall load–displacement response in the x -direction during the equi-biaxial loading/unloading. The points of the snapshots of Figure 3a–e are also labeled along the curves. As can be seen, similar to the case of uniaxial compression discussed in Section 3.1, the equi-biaxial loading and unloading responses also followed the same path. The entire history of the reversible

deformation involving mechanical instabilities could thus be captured by our modeling approach in a straightforward manner.

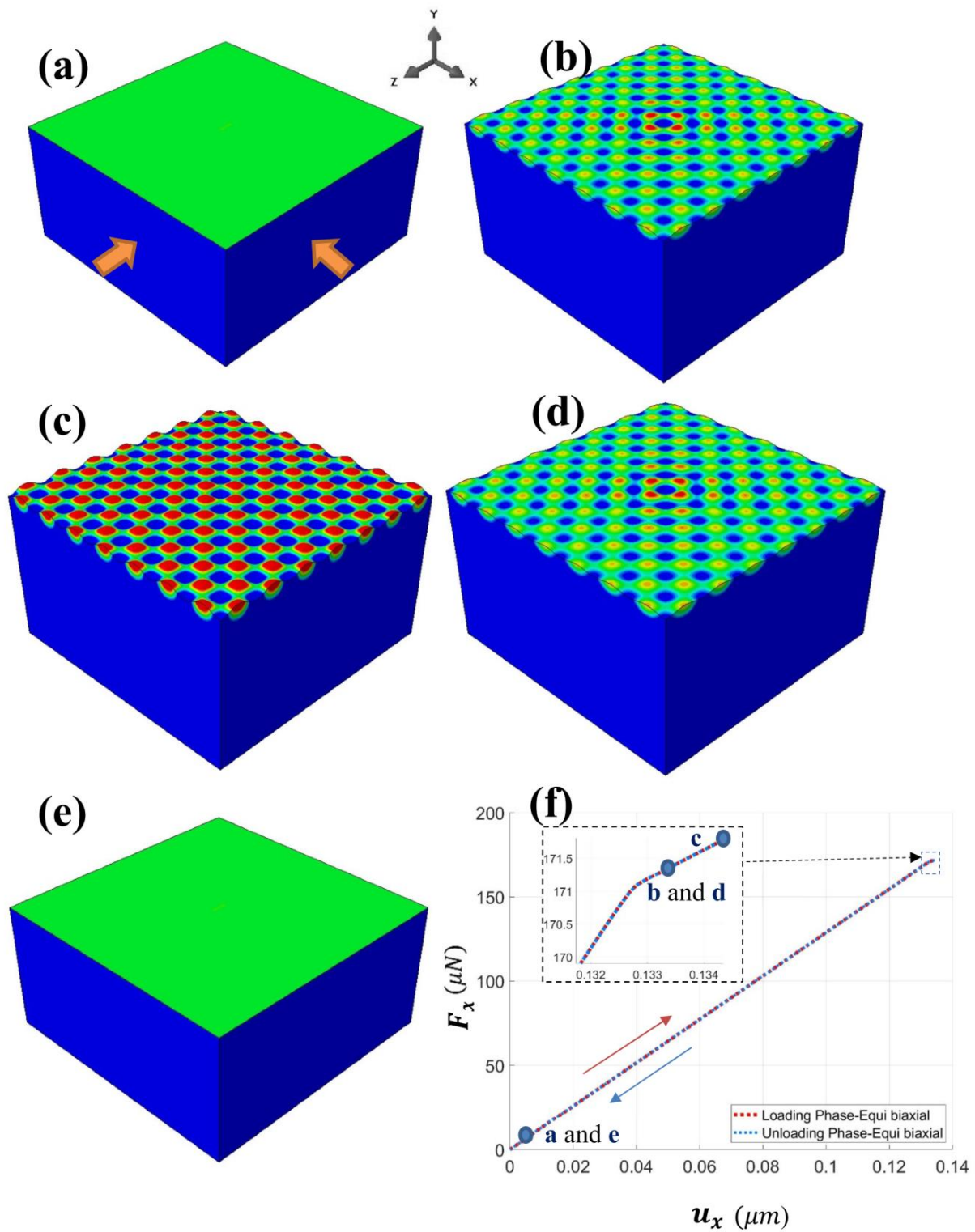


Figure 3. Snapshots of simulated surface pattern during the loading phase (from (a–c)) and unloading phase (from (c–e)) of the equi-biaxial compression. (f) The overall load–displacement response for the entire deformation history, with the five stages (a–e) labeled along the curves.

3.3. Simultaneous vs. Sequential Loading

In this section, we compare the surface wrinkling patterns based on simultaneous and sequential loadings leading to the same final state of strains. Consider the case of equi-biaxial compression ($BR = 1$) where the target applied strain corresponds to $e_{xx}/e_{cr} \cong 1.03$ (maximum equi-biaxial displacement of $-0.138 \mu\text{m}$). The result of simultaneous loading is presented in Section 3.2. For a sequential loading, a uniform compression along the x -direction only was first applied, which was followed by a second phase of compression along the z -direction.

At the end of the second phase, $u_x = u_z = -0.138 \mu\text{m}$, which is the same as the case presented in Section 3.2. Figure 4a,b show the surface patterns at the end of each phase of the sequential loading. After phase 1, the surface remained flat because the applied displacement had not reached the critical value for uniaxial compression (see Figure 2f). At the end of phase 2, a checkerboard-like pattern appeared, but it was not of the exact square type, as can be seen from the different numbers of waves along the x - and z -directions.

Figure 4c,d show the top views of the surface patterns resulting from the simultaneous and sequential loadings, respectively, for a direct comparison. Despite the same end state of equi-biaxial compressive strains, the sequential loading led to a non-square checkerboard pattern, as opposed to the perfect square checkerboard pattern obtained with the simultaneous loading. It is apparent that the mechanical instability rendered the simple elastic deformation history-dependent. Such a dependence may be attributed to the change in the domain geometry after the first phase of compression along the x -direction. The longer span in the z -direction at the end of the first phase (or equivalently, at the start of the second phase), coupled with the fixed constraint maintained in the x -direction during the second phase, contributed to the formation of a rectangular type of checkerboard pattern. The present finding also brings about the possibilities for creating variants of wrinkle patterns controlled by the deformation path, which is worthy of further theoretical and experimental investigations.

3.4. Simultaneous vs. Sequential Loading: Further Instability Modes

The history dependency of the surface wrinkling patterns was not limited to the primary bifurcation mode. It was even more apparent as the deformation extended into the post-instability regime. For demonstration, consider the same problem in Figure 4, but the specimen was now subjected to a higher maximum equi-biaxial target displacement of $-0.51 \mu\text{m}$ (which corresponds to $e_{xx}/e_{cr} \cong 3.80$). Figure 5a,b show the final surface states for the simultaneous and sequential loadings, respectively, with very different wrinkle patterns. Their temporal developments for the cases of simultaneous and sequential loading are presented in Figure 5c,d, respectively. Under simultaneous loading (Figure 5c), the primary instability mode was the square checkerboard, which, with further straining, eventually evolved into a labyrinth pattern. Under sequential loading (Figure 5d), the primary instability was the 1D mode, since phase 1 involved only a uniaxial compression applied in the x -direction. During phase 2, the superposition of z -compression led to a herringbone-like structure, which eventually became a labyrinth. It was again observed that the same final strain states in Figure 5c,d via different deformation histories, had distinctly different surface patterns.

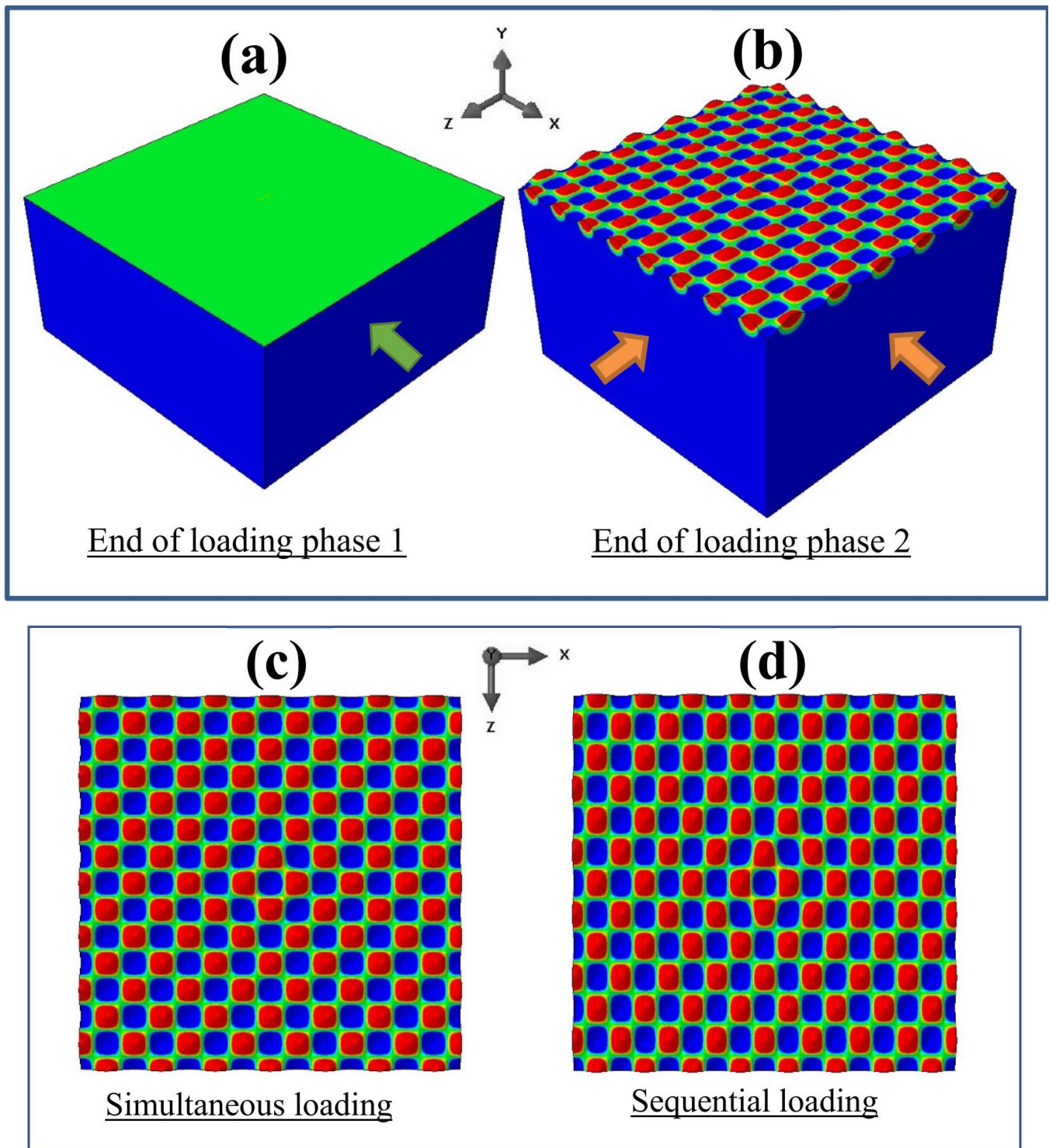


Figure 4. (a,b): Snapshots of simulated surface patterns during a sequential loading at (a) the end of phase 1 and (b) at the end of phase 2. (c) Square checkerboard wrinkling pattern obtained from a simultaneous loading. (d) Non-square checkerboard wrinkling pattern obtained from a sequential loading.

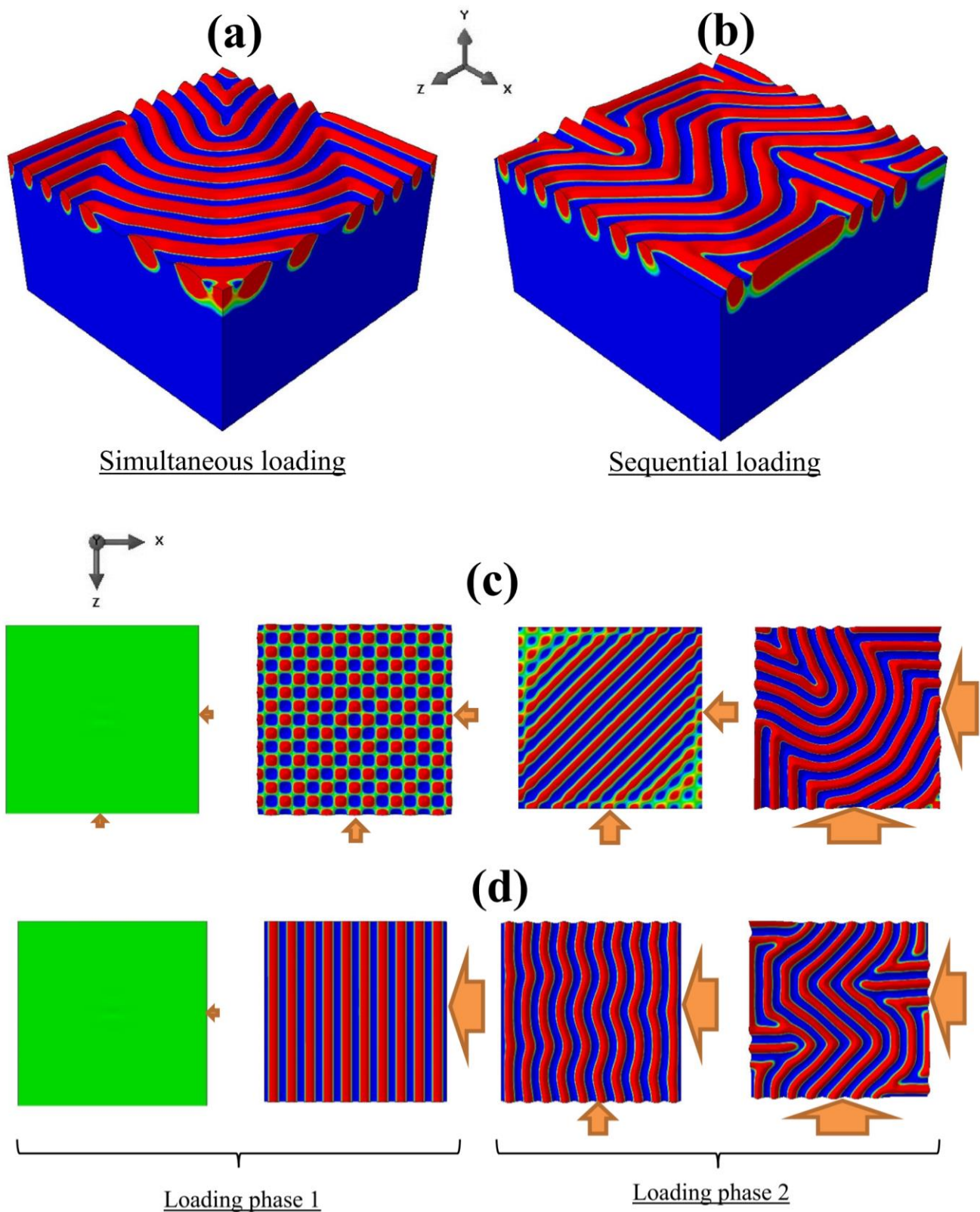


Figure 5. Snapshots of the simulated wrinkling patterns for $BR = 1.0$ and $e_{xx}/e_{cr} \cong 3.80$, for the cases of (a,c): simultaneous loading and (b,d): sequential loading. The wrinkle configurations at the end of each simulation are presented in (a,b). The evolution of the wrinkling patterns (starting from a flat surface until reaching the target strains) are shown in (c,d). The directions of the applied displacements are also schematically shown.

4. Discussion

The results presented in Sections 3.3 and 3.4 clearly demonstrated the path dependence of surface instability, even when the materials are treated as linearly elastic. It is worthy of note that except for very rare cases [45], a true square-checkerboard wrinkling configuration has never been obtained in real experimentation [15]. Various potential reasons, such as a curved initial geometry and a special constitutive behavior of the materials, have been proposed [15,25,30,39,46]. The finding presented in Figure 4 of this work, under equi-biaxial compression, points to yet another possibility, i.e., that deviations from the true simultaneous uniform loading along the two directions (e.g., slight interruptions of loading continuity in any one direction) may create a favorable condition for the non-square-type checkerboard pattern.

Aside from the uniaxial and equi-biaxial loadings considered above, one can explore a non-equi-biaxial loading with the BR value falling between 0 and 1. Here, we will discuss the case of $BR = 0.7$. Three sets of simulations with the maximum applied displacements of $u_x = -0.51 \mu\text{m}$ and $u_z = -0.357 \mu\text{m}$, corresponding to a final deformation of $e_{xx}/e_{cr} \cong 3.50$ ($e_{cr} = 0.0026$), were first considered: (i) simultaneous loading along the x - and z -directions, (ii) two-phase sequential loading with x -compression applied first and z -compression applied afterward, (iii) two-phase sequential loading with z -compression applied first and x -compression afterward. In the following, the two sequential cases (ii) and (iii) are also referred to as “sequential loading 1” and “sequential loading 2,” respectively. The numerical results are shown in Figure 6, with Figure 6a–c displaying the final wrinkle patterns at the end of simultaneous loading, sequential loading 1, and sequential loading 2, respectively. As can be seen, different wrinkle patterns were obtained with different deformation paths, while the loading biaxiality was identical ($BR = 0.7$) at the end of each simulation.

The temporal evolutions of instability are presented in Figure 6d for simultaneous loading, Figure 6e for sequential loading 1, and Figure 6f for sequential loading 2, all from a flat pre-instability state well into post-instability. Under simultaneous loading at $BR = 0.7$ (Figure 6d), the primary 1D instability mode transformed into a herringbone and later a labyrinth pattern, which is consistent with our previous studies and other experimental/analytical investigations reported in the literature [15,25,39,44,47]. A qualitatively similar evolution path was observed for the case of sequential loading 1 (Figure 6e), where the x -compression was applied first (during loading phase 1). It should be noted that, although the evolution paths in Figure 6d,e are similar, the actual herringbone and labyrinth surface patterns were different. This difference can be attributed to the more prominent sinusoidal wave amplitude in the case of sequential loading 1 at the end of loading phase 1, since the maximum applied displacement of $u_x = -0.51 \mu\text{m}$ was already reached, while in the case of simultaneous loading, the mode transformation from a 1D to a herringbone pattern happened much earlier. Sequential loading 2, where the z -compression was applied first, triggered the formation of 1D wrinkles in the perpendicular direction (Figure 6f). Note that, with $BR = 0.7$, the applied displacement at the end of the loading phase 1 was $u_z = -0.357 \mu\text{m}$, which rendered yet another sinusoidal wave amplitude. A subsequent x -compression during the loading phase 2 therefore led to different forms of herringbone and then labyrinth configurations. It is worth noting that the wrinkle amplitude was directly proportional to the intensity of the applied strain, as documented in many analytical/experimental/numerical studies in the literature [14,15,39].

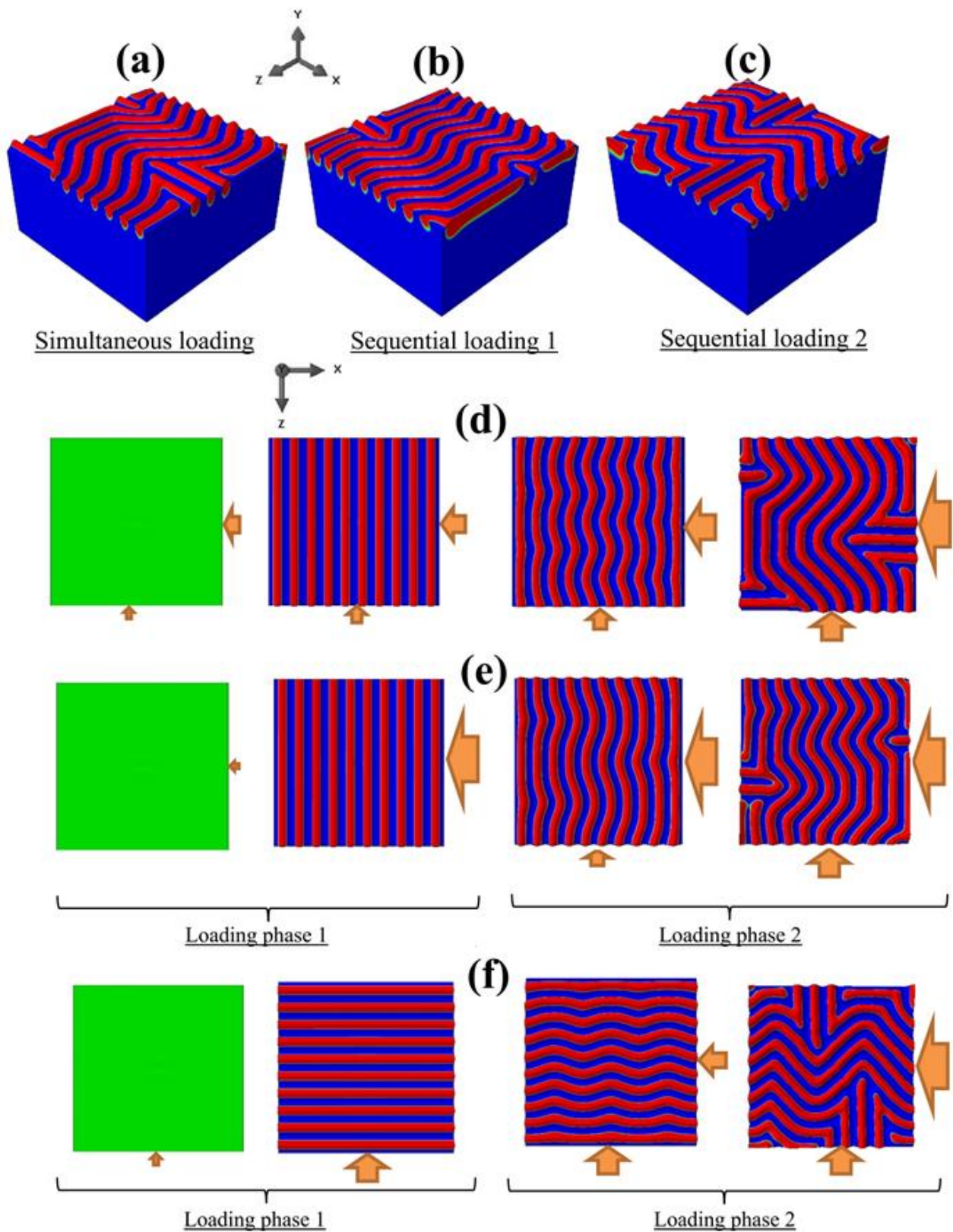


Figure 6. Snapshots of the simulated wrinkling patterns for $BR = 0.7$ and $e_{xx}/e_{cr} \cong 3.50$, for the case of (a,d): simultaneous loading, (b,e): sequential loading 1, and (c,f): sequential loading 2. The wrinkling configurations at the end of each simulation are presented in (a–c). The evolution of the wrinkling patterns (starting from a flat surface until reaching the target strains) are shown in (d–f). The directions of the applied displacements are also schematically shown.

Figure 7 shows the results for another set of simulations at $BR = 0.7$, with a smaller target strain of $e_{xx}/e_{cr} \cong 1.96$. The maximum applied displacements were $u_x = -0.284 \mu\text{m}$ and $u_z = -0.199 \mu\text{m}$. The final wrinkling configurations are shown in Figure 7a–c, respectively, for the three cases of simultaneous loading, sequential loading 1 (x -compression first), and sequential loading 2 (z -compression first). The history-dependent wrinkle patterns are again evident. The deformation progression is shown in Figure 7d–f for the cases of simultaneous loading, sequential loading 1, and sequential loading 2, respectively. As in the case of Figure 6, simultaneous loading in Figure 7d and sequential loading 1 (x -compression first) in Figure 7e resulted in a similar kind of pattern evolution, but the detailed wrinkle morphologies were not the same. An interesting pattern evolution, however, was observed for sequential loading 2 in Figure 7f (with z -compression first). Here, the primary instability did not occur at the end of the loading phase 1. This was due to the smaller target strain; the maximum applied displacement ($u_z = -0.199 \mu\text{m}$) corresponded to the applied strain of $e_{zz} = -0.0036$, which was still below the critical strain of $e_{cr} \cong -0.0037$. At the beginning of the second loading phase, only a slight compression of u_x triggers the formation of 1D wrinkles in the original z -direction. With increasing x -compression, the bifurcation mode changed to herringbone and then labyrinth.

Figures 5–7 demonstrated that the history dependency is not limited to the primary form of instability. The development of further wrinkle patterns can be influenced by the sequence of loading and its extent. The findings in this work also suggest wide-ranging possibilities for generating variants of wrinkle patterns controlled by the deformation history for various functionalities, including optics [6], friction [46], wetting [5], antifouling [48], etc. It is also worthy of note that the embedded imperfection technique used here is particularly suited for multi-phase loading processes such as cyclic deformation. The need for re-defining imperfections in each loading phase can be completely avoided. Future studies may incorporate inelastic and/or damage constitutive behaviors to simulate complex cyclic deformation histories and fatigue damage of film–substrate systems.

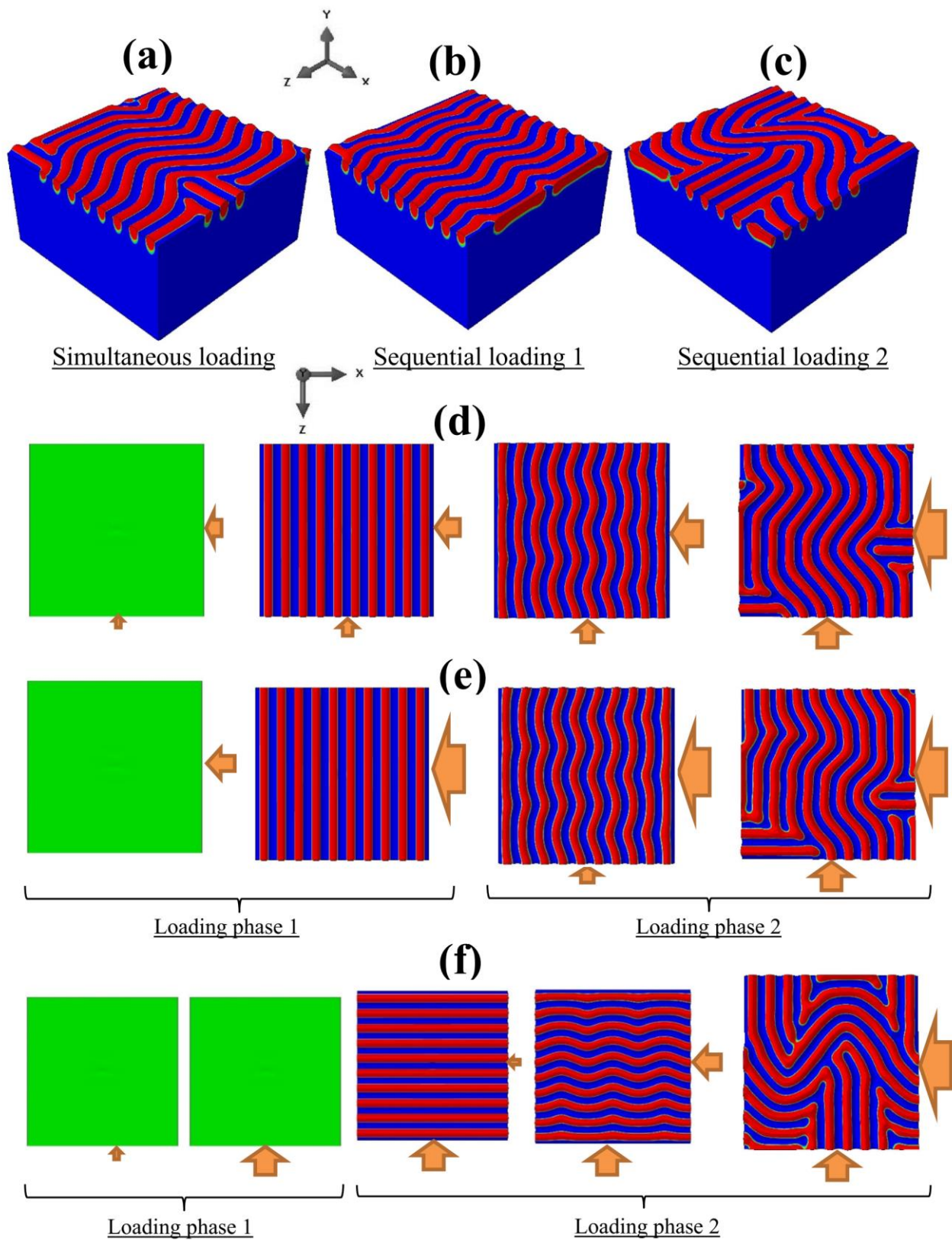


Figure 7. Snapshots of the simulated wrinkling patterns for $BR = 0.7$ and $e_{xx}/e_{cr} \cong 1.96$, for the case of (a,d): simultaneous loading, (b,e): sequential loading 1, and (c,f): sequential loading 2. The wrinkling configurations at the end of each simulation are presented in (a–c). The evolution of the wrinkling patterns (starting from a flat surface until reaching the target strains) are shown in (d–f). The directions of the applied displacements are also schematically shown.

5. Conclusions

This paper presents a numerical study on the formation of wrinkle patterns in thin film/compliant substrate material systems. The embedded imperfection approach was successfully applied for direct three-dimensional finite element simulations of surface wrinkling, accounting for loading/unloading and the effect of the deformation history. The modeling technique is robust and easy to implement and avoids the need of re-defining imperfections in each loading phase. Other salient findings and concluding remarks are summarized below.

- Within the linear elastic framework free of material damage, deformation instabilities in the form of surface wrinkles are recoverable under uniaxial and equi-biaxial loading and unloading.
- Using different deformation paths to reach the same equi-biaxial and non-equi-biaxial states, however, results in different wrinkle configurations.
- The history dependency is applicable to the primary instability mode as well as to subsequent transformations of wrinkle patterns.
- The history dependency also raises the possibilities of devising special loading sequences in actual experiments to achieve specific surface patterns.
- This study paves the way for future explorations involving complex deformation paths, inelastic deformation and damage, and cyclic responses.

Author Contributions: S.N. and Y.-L.S. conceived the idea for the research and contributed to writing the manuscript. S.N. performed the numerical simulations directed by Y.-L.S. All authors have read and agreed to the published version of the manuscript.

Funding: This work was partially supported by an endowment from the PNM Resources Foundation to Y.-L.S. as the PNM Chair for Renewable Energy Research.

Data Availability Statement: Not applicable.

Acknowledgments: The authors acknowledge the Center for Advanced Research Computing at the University of New Mexico, supported in part by the National Science Foundation, for providing the high-performance computing resources used in this work.

Conflicts of Interest: The authors declare no conflict of interest. The funder had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

References

1. Chung, J.Y.; Nolte, A.J.; Stafford, C.M. Surface wrinkling: A versatile platform for measuring thin-film properties. *Adv. Mater.* **2011**, *23*, 349–368. [[CrossRef](#)] [[PubMed](#)]
2. Khang, D.Y.; Rogers, J.A.; Lee, H.H. Mechanical buckling: Mechanics, metrology, and stretchable electronics. *Adv. Funct. Mater.* **2009**, *19*, 1526–1536. [[CrossRef](#)]
3. Wang, Q.; Zhao, X. Beyond wrinkles: Multimodal surface instabilities for multifunctional patterning. *MRS Bull.* **2016**, *41*, 115–122. [[CrossRef](#)]
4. Bangsund, J.S.; Fielitz, T.R.; Steiner, T.J.; Shi, K.; Van Sambeek, J.R.; Clark, C.P.; Holmes, R.J. Formation of aligned periodic patterns during the crystallization of organic semiconductor thin films. *Nat. Mater.* **2019**, *18*, 725. [[CrossRef](#)] [[PubMed](#)]
5. Lee, W.-K.; Jung, W.-B.; Nagel, S.R.; Odom, T.W. Stretchable superhydrophobicity from monolithic, three-dimensional hierarchical wrinkles. *Nano Lett.* **2016**, *16*, 3774–3779. [[CrossRef](#)]
6. Wu, K.; Sun, Y.; Yuan, H.; Zhang, J.; Liu, G.; Sun, J. Harnessing dynamic wrinkling surfaces for smart displays. *Nano Lett.* **2020**, *20*, 4129–4135. [[CrossRef](#)]
7. Nikraves, S.; Ryu, D.; Shen, Y.-L. Direct numerical simulation of buckling instability of thin films on a compliant substrate. *Adv. Mech. Eng.* **2019**, *11*, 1687814019840470. [[CrossRef](#)]
8. Ryu, D.; Mongare, A. Corrugated Photoactive Thin Films for Flexible Strain Sensor. *Materials* **2018**, *11*, 1970. [[CrossRef](#)]
9. Lipomi, D.J.; Tee, B.C.K.; Vosgueritchian, M.; Bao, Z. Stretchable organic solar cells. *Adv. Mater.* **2011**, *23*, 1771–1775. [[CrossRef](#)]
10. Kim, J.B.; Kim, P.; Pégard, N.C.; Oh, S.J.; Kagan, C.R.; Fleischer, J.W.; Stone, H.A.; Loo, Y.-L. Wrinkles and deep folds as photonic structures in photovoltaics. *Nat. Photonics* **2012**, *6*, 327–332. [[CrossRef](#)]
11. Dimmock, R.L.; Wang, X.; Fu, Y.; El Haj, A.J.; Yang, Y. Biomedical applications of wrinkling polymers. *Recent Prog. Mater.* **2020**, *2*, 1–31. [[CrossRef](#)]

12. Watanabe, M.; Tazawa, H.; Suganuma, K. Preparation of drug microparticles with a narrow size distribution using regular dimples induced by buckling instability. *Appl. Surf. Sci.* **2021**, *563*, 150251. [\[CrossRef\]](#)
13. Gabardo, C.M.; Yang, J.; Smith, N.J.; Adams-McGavin, R.C.; Soleymani, L. Programmable wrinkling of self-assembled nanoparticle films on shape memory polymers. *ACS Nano* **2016**, *10*, 8829–8836. [\[CrossRef\]](#) [\[PubMed\]](#)
14. Volynskii, A.; Bazhenov, S.; Lebedeva, O.; Bakeev, N. Mechanical buckling instability of thin coatings deposited on soft polymer substrates. *J. Mater. Sci.* **2000**, *35*, 547–554. [\[CrossRef\]](#)
15. Cai, S.; Breid, D.; Crosby, A.J.; Suo, Z.; Hutchinson, J.W. Periodic patterns and energy states of buckled films on compliant substrates. *J. Mech. Phys. Solids* **2011**, *59*, 1094–1114. [\[CrossRef\]](#)
16. Chen, X.; Hutchinson, J.W. Herringbone buckling patterns of compressed thin films on compliant substrates. *J. Appl. Mech.* **2004**, *71*, 597–603. [\[CrossRef\]](#)
17. Song, J.; Jiang, H.; Choi, W.; Khang, D.-Y.; Huang, Y.; Rogers, J.A. An analytical study of two-dimensional buckling of thin films on compliant substrates. *J. Appl. Phys.* **2008**, *103*, 014303. [\[CrossRef\]](#)
18. Sui, J.; Chen, J.; Zhang, X.; Nie, G.; Zhang, T. Symplectic analysis of wrinkles in elastic layers with graded stiffnesses. *J. Appl. Mech.* **2019**, *86*, 011008. [\[CrossRef\]](#)
19. Huck, W.T.; Bowden, N.; Onck, P.; Pardo, T.; Hutchinson, J.W.; Whitesides, G.M. Ordering of spontaneously formed buckles on planar surfaces. *Langmuir* **2000**, *16*, 3497–3501. [\[CrossRef\]](#)
20. Cao, Y.; Hutchinson, J.W. From wrinkles to creases in elastomers: The instability and imperfection-sensitivity of wrinkling. *Proc. R. Soc. A* **2012**, *468*, 94–115. [\[CrossRef\]](#)
21. Cao, Y.-P.; Zheng, X.-P.; Jia, F.; Feng, X.-Q. Wrinkling and creasing of a compressed elastoplastic film resting on a soft substrate. *Comput. Mater. Sci.* **2012**, *57*, 111–117. [\[CrossRef\]](#)
22. Saha, S.K. Sensitivity of the mode locking phenomenon to geometric imperfections during wrinkling of supported thin films. *Int. J. Solids Struct.* **2017**, *109*, 166–179. [\[CrossRef\]](#)
23. Mei, H.; Landis, C.M.; Huang, R. Concomitant wrinkling and buckle-delamination of elastic thin films on compliant substrates. *Mech. Mater.* **2011**, *43*, 627–642. [\[CrossRef\]](#)
24. Miyoshi, H.; Matsubara, S.; Okumura, D. Bifurcation and deformation during the evolution of periodic patterns on a gel film bonded to a soft substrate. *J. Mech. Phys. Solids* **2020**, *148*, 104272. [\[CrossRef\]](#)
25. Huang, X.; Li, B.; Hong, W.; Cao, Y.-P.; Feng, X.-Q. Effects of tension–compression asymmetry on the surface wrinkling of film–substrate systems. *J. Mech. Phys. Solids* **2016**, *94*, 88–104. [\[CrossRef\]](#)
26. Xu, F.; Potier-Ferry, M.; Belouettar, S.; Cong, Y. 3D finite element modeling for instabilities in thin films on soft substrates. *Int. J. Solids Struct.* **2014**, *51*, 3619–3632. [\[CrossRef\]](#)
27. Xu, F.; Koutsawa, Y.; Potier-Ferry, M.; Belouettar, S. Instabilities in thin films on hyperelastic substrates by 3D finite elements. *Int. J. Solids Struct.* **2015**, *69*, 71–85. [\[CrossRef\]](#)
28. Okumura, D.; Sugiura, J.; Tanaka, H.; Shibutani, Y. Buckling and postbuckling of etching-induced wiggling in a bilayer structure with intrinsic compressive stress. *Int. J. Mech. Sci.* **2018**, *141*, 78–88. [\[CrossRef\]](#)
29. Hu, H.; Cao, D.; Cao, Z.; Li, S. Experimental and numerical investigations of wrinkle effect on failure behavior of curved composite laminates. *Compos. Struct.* **2021**, *261*, 113541. [\[CrossRef\]](#)
30. Zhao, Y.; Zhu, H.; Jiang, C.; Cao, Y.; Feng, X.-Q. Wrinkling pattern evolution on curved surfaces. *J. Mech. Phys. Solids* **2020**, *135*, 103798. [\[CrossRef\]](#)
31. Stoop, N.; Wittel, F.K.; Amar, M.B.; Müller, M.M.; Herrmann, H.J. Self-contact and instabilities in the anisotropic growth of elastic membranes. *Phys. Rev. Lett.* **2010**, *105*, 068101. [\[CrossRef\]](#) [\[PubMed\]](#)
32. Li, B.; Jia, F.; Cao, Y.-P.; Feng, X.-Q.; Gao, H. Surface wrinkling patterns on a core-shell soft sphere. *Phys. Rev. Lett.* **2011**, *106*, 234301. [\[CrossRef\]](#) [\[PubMed\]](#)
33. Ebrahimi, S. Mechanical Behavior of Materials at Multiscale Peridynamic Theory and Learning-Based Approaches. Ph.D. Thesis, University of California, Berkeley, CA, USA, 2020.
34. Laurien, M.; Javili, A.; Steinmann, P. Nonlocal wrinkling instabilities in bilayered systems using peridynamics. *Comput. Mech.* **2021**, *68*, 1023–1037. [\[CrossRef\]](#)
35. Breid, D.; Crosby, A.J. Effect of stress state on wrinkle morphology. *Soft Matter* **2011**, *7*, 4490–4496. [\[CrossRef\]](#)
36. Nikraves, S.; Ryu, D.; Shen, Y.-L. Surface Instability of Composite Thin Films on Compliant Substrates: Direct Simulation Approach. *Front. Mater.* **2019**, *6*, 214. [\[CrossRef\]](#)
37. Nikraves, S.; Ryu, D.; Shen, Y.-L. Instabilities of Thin Films on a Compliant Substrate: Direct Numerical Simulations from Surface Wrinkling to Global Buckling. *Sci. Rep.* **2020**, *10*, 5728. [\[CrossRef\]](#)
38. Nikraves, S.; Ryu, D.; Shen, Y.-L. Instability driven surface patterns: Insights from direct three-dimensional finite element simulations. *Extrem. Mech. Lett.* **2020**, *39*, 100779. [\[CrossRef\]](#)
39. Nikraves, S.; Ryu, D.; Shen, Y.-L. Direct numerical simulations of three-dimensional surface instability patterns in thin film-compliant substrate structures. *Sci. Rep.* **2021**, *11*, 16449. [\[CrossRef\]](#)
40. Tahk, D.; Lee, H.H.; Khang, D.-Y. Elastic moduli of organic electronic materials by the buckling method. *Macromolecules* **2009**, *42*, 7079–7083. [\[CrossRef\]](#)
41. Nikraves, S.; Ryu, D.; Shen, Y.-L. Surface Wrinkling versus Global Buckling Instabilities in Thin Film-Substrate Systems under Biaxial Loading: Direct 3D Numerical Simulations. *Adv. Theory Simul.* **2022**, *5*, 2200183. [\[CrossRef\]](#)

42. Shen, Y.-L. *Constrained Deformation of Materials: Devices, Heterogeneous Structures and Thermo-Mechanical Modeling*; Springer Science & Business Media: New York, NY, USA, 2010.
43. Allen, H.G. Analysis and design of structural sandwich panels. In *Analysis and Design of Structural Sandwich Panels*, 1st ed.; Pergamon Press Ltd., Headinton Hill Hall: London, UK, 1969; pp. 76–97.
44. Huang, Z.; Hong, W.; Suo, Z. Nonlinear analyses of wrinkles in a film bonded to a compliant substrate. *J. Mech. Phys. Solids* **2005**, *53*, 2101–2118. [[CrossRef](#)]
45. Watanabe, M. Shrink-induced striped pattern on a thin gold film and switching of its orientation. *J. Appl. Polym. Sci.* **2006**, *101*, 2040–2044. [[CrossRef](#)]
46. Yuan, H.; Wu, K.; Zhang, J.; Wang, Y.; Liu, G.; Sun, J. Curvature-Controlled Wrinkling Surfaces for Friction. *Adv. Mater.* **2019**, *31*, 1900933. [[CrossRef](#)]
47. Huang, Z.; Hong, W.; Suo, Z. Evolution of wrinkles in hard films on soft substrates. *Phys. Rev. E* **2004**, *70*, 030601. [[CrossRef](#)] [[PubMed](#)]
48. Epstein, A.K.; Hong, D.; Kim, P.; Aizenberg, J. Biofilm attachment reduction on bioinspired, dynamic, micro-wrinkling surfaces. *N. J. Phys.* **2013**, *15*, 095018. [[CrossRef](#)]