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Surface effect on the buckling of piezoelectric nanofilms

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Abstract
A sandwich-plate model is developed to account for the effect of surface layers on the buckling of piezoelectric nanofilms (PNFs) due to an electrical voltage. The physical mechanisms of the surface effects are investigated and the contributions to the resultant effect are evaluated for surface piezoelectricity, surface stress and surface elasticity. It is found that the surface effect originates primarily from the residual surface stresses and enhanced piezoelectric coefficient due to the surface piezoelectricity. Its influence on the critical buckling voltage of a PNF depends sensitively on the thickness, the length-to-thickness ratio and the nature of residual surface stress. In addition, the intrinsic buckling may occur for a thin PNF where the relatively strong residual surface compression is achieved.

1. Introduction
Piezoelectric nanostructures (PNs) have been synthesized in various configurations [1, 2], e.g. nanowires, nanofilms and nanorings. These smart nanomaterials exhibit enhanced piezoelectric effect, excellent resilience and the unique coupling between piezoelectric and semiconducting properties [1–3]. The distinct features make PNs promising for a wide range of applications in nanotechnology, such as nanosensors/transducers, nanogenerators, nanoresonators, diodes and piezoelectric field-effect transistors. The piezoelectric responses of PNs have thus become a current topic of great interest in recent research [4–13], where continuum piezoelectric theory was efficiently used as a cost-effective technique.

It is well-known that miniaturization of materials into a nanoscale size will significantly increase the surface-to-volume ratio. It thus may substantially improve the effect of the surface layers. This presumption has already been confirmed for elastic nanomaterials where significant surface effect leads to size-dependent elastic properties [14, 15] and the vibration and buckling behaviour distinct from those of their bulk counterparts [16, 17]. Recently, the size-dependent piezoelectric properties were reported for PNs in experiments [18] and simulations [19–21]. This infers that in addition to the surface stresses and surface elasticity, the surface piezoelectricity could also play a role in controlling the structural responses of PNs. However, most early studies of PNs [4–8] were based on the classical piezoelectric theory without considering the surface layers. Progress was made in a recent work by Wang and Feng [9], where an elastic surface layer was introduced. The effect of piezoelectric surface layers was first examined by Huang and Yu [10] for the static deformation of a piezoelectric nano-annular. The idea was further extended by Yan and Jiang [11, 12] to the study of the bending, buckling and vibration of piezoelectric nanobeams. Very recently, based on the one-dimensional beam theory Li et al [13] examined the effect of piezoelectric surface on the wrinkling of piezoelectric nanofilms (PNFs). Nevertheless PNFs as two-dimensional (2D) piezoelectric nanomaterials have not yet been studied so far due to lack of a reliable 2D model for PNFs. In particular, the physical mechanisms of the surface effect and the role of the surface stress, surface elasticity and surface piezoelectricity have not been discussed in detail.

To address these fundamental issues, this work aims to study the surface effect on the structural responses of 2D PNFs. In particular, the buckling induced by an electrical voltage is considered as it forms one of the major concerns for piezoelectric thin structures such as PNFs. A sandwich-plate model is first developed for the 2D piezoelectric nanomaterials by incorporating the surface effect and piezoelectric effect into
An isotropic rectangular PNF with width $2b$, length $a$, and constant thickness $h$ is illustrated in figure 1. As seen from the illustration Cartesian coordinate system $x_0$ ($\alpha = 1, 2, 3$) is used for the PNF, where the $x_1x_3$ plane coincides with its undeformed midplane and $x_1$ is in the direction of its thickness. The usual summation convention for repeated indices will be adopted with Greek indices ranging from 1 to 3 and Latin ones taking the values of 1 and 2. A comma represents differentiation with respect to the superscript index.

To account for the effect of the surface layers a PNF is considered as a composite plate comprising a core layer that is sandwiched between top and bottom thin surface layers (figure 1). The core plate has a constant thickness $h$ (equal to the thickness of the PNF) and the same material properties as those of the bulk material. On the other hand, the two surface layers are treated as 2D plates with bending stiffness and in-plane extensional stiffness but negligible thickness. Residual stresses and the elastic, piezoelectric and dielectric properties different from those of the core layer are considered for the surface layers. In addition, a perfect bond is achieved between the core plate and the two surface layers, i.e. no slipping occurs between the two adjacent components, and the displacements are continuous in the sandwich plate. Here we first use Mindlin plate theory accounting for the effect of the shear deformation to derive the formulations. Following Mindlin’s assumption the displacement field $u_\alpha$ at a point of the film (including the surface layers) can be represented by

$$u_\alpha = u_\alpha^0 - x_3 \psi_i, \quad u_3 = u_3^0 \equiv w(x_1, t)$$

where $u_\alpha^0$ is the displacement components of the midplane along the $x_\alpha$ direction, and $\psi_i$ is the rotation angle of a normal line due to plate bending with respect to $x_1$ coordinates. In particular, $\psi_i$ can be approximated by $\psi_i = w_{i,3}$ for a Kirchhoff plate. Accordingly, the strains at an arbitrary point of PNFs can be written as

$$\varepsilon_{ij} = \left( u_{i,j} + u_{j,i} \right) / 2 = \gamma_{ij} + x_3 ( \phi_{i,j} + \phi_{j,i} ) / 2,$$  \hspace{1cm} (2)

$$\varepsilon_{i3} = \left( u_{i,3} + u_{3,i} \right) / 2 = (w_{i,3} - \phi_i) / 2$$  \hspace{1cm} (3)

where $\gamma_{ij} = (u_{i,j}^0 + u_{j,i}^0) / 2$ are the midplane strains describing the membrane deformations. The electric field is assumed to exist only in the $x_3$ direction, and related to the electric potential $\phi$ by

$$E_3 = -\phi_3.$$  \hspace{1cm} (4)

For the core layer of PNFs the stresses $\sigma_{ij}$ and the electric displacement $D_3$ can be written as

$$\sigma_{ij} = c_{ijkl} e_{kl} - e_{ij3} E_3$$  \hspace{1cm} (5)

$$D_3 = e_{ij3} E_3 + k_{33} E_3$$  \hspace{1cm} (6)

where $c_{ijkl}$, $e_{ij3}$ and $k_{33}$ are, respectively, the elastic, piezoelectric and dielectric constants for the bulk material. It is understood that the thickness of the surface layers is usually orders of magnitude smaller than the thickness of nanomaterials [22]. They are thus considered as two-dimensional (2D) surfaces (without thickness) in this work as well as previous studies of piezoelectric nanomaterials [10–12]. For such 2D surfaces the constitutive relations read [10–12]

$$\sigma_{ij} = \sigma_{ij}^0 + c_{ij3} w_{i,3} + e_{ij3} E_3$$  \hspace{1cm} (7)

where $\sigma_{ij}^0$ are membrane forces per unit length (or surface stresses), $\sigma_{ij}^0$ are residual membrane forces per unit length (or residual surface stresses), $c_{ijkl}$ are the in-plane stiffnesses and $e_{ij3}$ are the piezoelectric constants relating electric field and the membrane forces. In the absence of free electric charges, the electrostatic equilibrium condition requires that

$$D_{3,3} = 0.$$  \hspace{1cm} (8)

Combining equations (1), (2), (4) and (8), and the electrical boundary conditions $\phi(-h/2) = 0$ and $\phi(h/2) = V$, the electric potential distribution is determined as

$$\phi = -\frac{1}{2k_{33}} e_{ij3} \psi_{ij} x_3^2 - \frac{h^2}{4} + \frac{V}{h} x_3 + \frac{V}{2}.$$  \hspace{1cm} (9)

Substituting equations (2), (3) and (9) into equations (5) and (7) yields the stresses in the core and surface layers

$$\sigma_{ij} = c_{ijkl} \gamma_{kl} - x_3 \tilde{c}_{ijkl} \psi_{kl} + e_{ij3} V / h$$  \hspace{1cm} (10)

$$\sigma_{ij} = c_{ijkl} ( w_{i,j} - \phi_i )$$  \hspace{1cm} (11)

$$\sigma_{ij} = \sigma_{ij}^0 + c_{ij3} w_{i,3} + e_{ij3} V / h$$  \hspace{1cm} (12)

where $\tilde{c}_{ijkl} = c_{ijkl} + e_{ij3} e_{kl3} / k_{33}$ and $c_{ij3}^l = c_{ij3} + e_{ij3} e_{kl3}^l / k_{33}$. The stress resultant $N_{i\alpha}$ and stress moment $M_{ij}$ can be calculated by

$$N_{ij} = \int_{-h/2}^{h/2} \sigma_{ij} \, dx_3 + (\sigma_{ij}^{i*} + \sigma_{ij}^{j*})$$

$$= K_{ijkl} \gamma_{kl} + e_{ij3} V + 2 \left( \sigma_{ij}^{i*} + \sigma_{ij}^{j*} V / h \right)$$  \hspace{1cm} (13)
\[ N_{i3} = k \int_{-h/2}^{h/2} \sigma_{i3} \, dx_3 = A \kappa c_{13j3} (w_{i,j} - \phi_j) \]  
(14)

\[ M_{ij} = \int_{-h/2}^{h/2} x_3 \sigma_{ij} \, dx_3 + \frac{h}{2} (\sigma_{ij}^{+} - \sigma_{ij}^{-}) = -D_{ijkl} \psi_{k,l}. \]  
(15)

Here \( \sigma_{ij}^{+} = \sigma_{ij}^{-} \) are the stresses on the top and bottom surface layers, \( k \) is the shear correction factor which usually has the value 5/6 [23] and \( D_{ijkl} \) and \( K_{ijkl} \) are the equivalent bending stiffness and in-plane extensional stiffness of the PNF, which are defined by

\[ D_{ijkl} = \left( c_{ijkl} + e_{ij3} e_{kl3} / k_{33} \right) I + \left( c_{ijkl}^{'} + e_{ij3}^{'} e_{kl3}^{'} / k_{33} \right) \frac{h^2}{2} \]  
(16)

\[ K_{ijkl} = A c_{ijkl} + 2 c_{ijkl}^{'} \]  
(17)

where \( (A, I) = \int_{-h/2}^{h/2} (1, x_1^2) \, dx_3 = (h, h^3/12) \). It is seen in equations (16) and (17) that both surface piezoelectricity and surface elasticity contribute to the bending stiffness of a PNF while only the surface elasticity affects its in-plane extensional stiffness. On the other hand, the residual surface stress has no effect on the structural stiffnesses of a PNF. It only acts as in-plane stresses in equation (13), where the equivalent in-plane stresses can also be found due to the surface piezoelectricity.

In Mindlin’s plate model, the equilibrium equation of a PNF is given as [23]

\[ M_{ij,j} - N_{i3} = 0 \]  
(18)

\[ N_{i3,i} + N_{ij} w_{ij,j} = 0. \]  
(19)

Eliminating \( N_{i3} \) we can get the following equilibrium equation for a Kirchhoff plate:

\[ M_{ij,j} + N_{ij} w_{ij,j} = 0. \]  
(20)

Substituting equations (13)–(15) into equations (18) and (19) yields the equilibrium equation of PNFs as Mindlin plates

\[ D_{ijkl} \psi_{k,l} + A \kappa c_{13j3} (w_{i,j} - \phi_j) = 0 \]  
(21)

\[ A \kappa c_{13j3} (w_{i,j} - \phi_j) + \left[ K_{ijkl} \gamma_{kl} + e_{ij3} V + 2 \left( \sigma_{ij}^{0} + e_{ij3} \frac{V}{h} \right) \right] w_{ij,j} = 0. \]  
(22)

Its counterpart for PNFs as Kirchhoff plates is given by

\[ D_{ijkl} w_{ij,k} - \left[ K_{ijkl} \gamma_{kl} + e_{ij3} V + 2 \left( \sigma_{ij}^{0} + e_{ij3} \frac{V}{h} \right) \right] w_{ij,j} = 0. \]  
(23)

To simplify our analysis, without losing generality, throughout the paper we consider PNFs with all boundaries simply supported. The solutions to equations (21) and (22) of such PNFs take the form

\[ \psi_1(x_1, x_2) = \Psi_{1mn} \cos \left( \frac{m \pi}{a} x_1 \right) \sin \left( \frac{n \pi}{b} x_2 \right) \]  
(24)

\[ \psi_2(x_1, x_2) = \Psi_{2mn} \sin \left( \frac{m \pi}{a} x_1 \right) \cos \left( \frac{n \pi}{b} x_2 \right) \]  
(25)

\[ w(x_1, x_2) = W_{mn} \sin \left( \frac{m \pi}{a} x_1 \right) \sin \left( \frac{n \pi}{b} x_2 \right) \]  
(26)

where \( \Psi_{1mn}, \Psi_{2mn} \) and \( W_{mn} \) are constants; and \( m \) and \( n \) are the half wave numbers. Substituting equations (24)–(26) into equations (21) and (22), and solving \( V \) from the resulting eigenvalue problem, we can obtain the buckling voltage \( V \) for a PNF as a function of \((m, n)\). The lowest value of \( V \) gives the critical buckling voltage \( V_{cr} \) of a PNF and associated \((m, n)\) describes the buckling mode. Similarly, after substituting equation (26) into equation (23) and solving \( V \) from the resulting equation we can get \( V_{cr} \) and corresponding \((m, n)\) for PNFs as Kirchhoff plates.

In figure 1, when the condition \( b > a \gg h \) is satisfied, a PNF becomes a beam-like structure where the transverse displacement \( w \) is a function of \( x_2 \) but independent of \( x_1 \). In this case it can be proved (not shown in this paper) that equation (23) will reduce to the governing equation for the vibration of a piezoelectric nanobeam given in [12].

3. Results and discussion

In this section the sandwich-plate model and the analysis methods developed in section 2 will be employed to conduct a quantitative study of the buckling behaviour of PNFs. PZT-5H (a lead zirconate titanate material) is selected as an example for piezoelectric materials. For the core layer of a PNF the bulk material properties are used, i.e. \( c_{11} = c_{22} = 126 \text{ GPa} \), \( c_{12} = 55 \text{ GPa} \), \( c_{44} = 35.3 \text{ GPa} \), \( c_{33} = -6.5 \text{ C m}^{-2} \) and \( k_{33} = 1.3 \times 10^{-8} \text{ C V}^{-1} \text{ m}^{-1} \) [24]. For the surface layers, the material properties can be obtained by atomistic calculations or experiments. However, the accurate values of surface piezoelectric and dielectric constants are not yet available due to lack of such work on piezoelectric nanomaterials. Thus the values estimated in [10] are used in this study as an approximation, i.e. \( c_{11}^{s} = c_{22}^{s} = 7.56 \text{ N m}^{-1} \) and \( c_{33}^{s} = -3 \times 10^{-8} \text{ C m}^{-1} \). Using an approximate ratio between different surface elastic constants [25], we obtain \( c_{11}^{s} = 5.23 \text{ N m}^{-1} \) and \( c_{22}^{s} = 1.26 \text{ N m}^{-1} \). In addition, \( \sigma_{ij}^{0} = 0 \) is assumed on the surface layers [26] and the effect of the small middle-plane strains on the elastic buckling is neglected by assuming \( \gamma_{ij} = 0 \).

3.1. Buckling behaviour of PNFs

Here let us first study the buckling behaviours of PNFs induced by an electrical voltage applied in their thickness direction. The residual surface stresses of PNFs are taken as \( \sigma_{ij}^{0} = 1 \text{ N m}^{-1} \). In figure 2 the critical electrical voltage \( V_{cr} \) is calculated against the aspect ratio \( a/h \) for the PNFs of a thickness \( h \) (a) 20 nm, (b) 200 nm and (c) 2 \( \mu \)m, respectively. The ratio \( a/b \) grows from 0.25 to 0.5 and to 1. To examine the effect of shear deformation, the results obtained based on Kirchhoff plate theory are also presented in figure 2 in comparison with those of Mindlin plate theory.

Figure 2(a) (for \( h = 20 \text{ nm} \)) shows that, with a given value of \( a/b \) the critical electric voltage \( V_{cr} \) decreases when the aspect ratio \( a/h \) grows (i.e. a PNF becomes thinner). For a fixed ratio \( a/h \) it declines with decreasing ratio \( a/b \).
change in the critical buckling voltage due to lack of reliable data in the literature. It, however, and finally becomes negligible at a small ratio (i.e. a PNF becomes longer). When the thickness $h$ increases to 200 nm and 2 $\mu$m, similar size-dependence of $V_{cr}$ is found in figures 2(b) and (c) but the value of $V_{cr}$ increases substantially in the process. For example, at $h = 20$ nm $V_{cr}$ falls in the range (0.4 V, 19 V) when $a/b = 1$ and $a/h$ varies between 5 and 50 (figure 2(a)). However, at $h = 200$ nm $V_{cr}$ rises to 2.8 to 251 V in the same range of $a/b$ and $a/h$ (figure 2(b)). Obviously, this $h$-dependence of $V_{cr}$ is a result of the enhanced bending stiffness of PNFs due to the increasing thickness $h$ (equation (16)). Here it is worth mentioning that $V_{cr}$ obtained in figure 2 always corresponds to the buckling mode $(m, n) = (1, 1)$ regardless of $a/h, a/b$ and $h$ considered.

In figure 2 the effect of shear deformation on the PNF buckling can be observed by comparing Mindlin plate theory considering shear deformation with Kirchhoff plate theory where the shear effect is neglected. It is noted that the shear deformation reduces the critical buckling voltage $V_{cr}$ significantly at a small ratio $a/h$. The effect then decreases with increasing $a/h$ and finally becomes negligible at $a/h \geq 20$. According to this comparison result, the sandwich-plate model based on the Mindlin plate theory will be used in general cases to account for the effect of the shear deformation, while the model based on Kirchoff theory will be used as an approximation in some special cases where $a/h \geq 20$ is satisfied. Here it is worth mentioning that when $a/h$ is small the critical buckling voltage could be even higher than the critical voltage of the electrical breakdown. In this particular case the electric breakdown will occur before buckling. This phenomenon has not been discussed in detail in this study due to lack of reliable data in the literature. It, however, needs to be taken into consideration when PNFs are put into a practical use.

### 3.2. Effect of surface layers

Next we shall study the effect of the surface layers on the buckling of PNFs. To this end we have calculated the relative change in the critical buckling voltage $(V_{cr} - V_{cr0})/V_{cr0}$ due to the surface effect, where $V_{cr0}$ is the critical buckling voltage obtained without considering the surface effect, i.e. $\sigma_{ijkl}^{(0)} = 0$, $\epsilon_{ijkl}^{(0)} = 0$ and $\sigma_{ijkl}^{(0)} = 0$. Previous studies [27, 28] showed that either residual surface tension or compression can be achieved for nanocrystals. Specifically, different surface stresses could alter the size-dependence of the surface effect on the buckling of PNFs. Thus, in this study we shall consider two cases: one is the PNFs made of PZT-5H where residual surface tension is found. The other is the PNFs made of a nanocrystal with residual surface compression. Here the objective is to capture the unique features of PNFs due to the surface compression. Thus the material properties of PZT-5H are still used in the second case.

The results obtained for PNFs with residual surface tension ($\sigma_{ij}^{(0)} = \sigma_{22}^{(0)} = 1$ N m$^{-1}$) are plotted versus $a/h$ in figure 3 with $a/b$ equal to (a) 0.25, (b) 0.5 and (c) 1, respectively, and the thickness $h$ varying from 20 nm, 200 nm to 2 $\mu$m. We can see from figure 3(a) that $(V_{cr} - V_{cr0})/V_{cr0}$ curves associated with different thicknesses $h$ intercept with the straight line $(V_{cr} - V_{cr0})/V_{cr0} = 0$ at exactly the same critical value of $a/h$, i.e. $(a/h)_{cr} = 24.1$ (see figure 3(a)). (1) At $a/h < (a/h)_{cr}$, i.e. PNFs are relatively thick, $(V_{cr} - V_{cr0})/V_{cr0}$ is negative suggesting that the existence of the surface layers reduces the critical buckling voltage $V_{cr}$. Such a surface effect measured by the absolute value of $(V_{cr} - V_{cr0})/V_{cr0}$ decreases when the aspect ratio $a/h$ rises to approach $(a/h)_{cr}$, i.e. PNFs become thinner. (2) When $a/h$ reaches the critical value $(a/h)_{cr}$, $(V_{cr} - V_{cr0})/V_{cr0}$ goes to zero and the surface effect vanishes. (3) Further raising $a/h$ to $a/h > (a/h)_{cr}$, i.e. PNFs are relatively thin, leads to positive $(V_{cr} - V_{cr0})/V_{cr0}$ which increases with growing $a/h$. Thus the surface layers of the relatively thin PNFs can raise the critical buckling voltage. This effect becomes more pronounced for thinner PNFs with a greater aspect ratio $a/h$.

From these results it follows that the critical value $(a/h)_{cr}$ is independent of the thickness $h$ and can classify PNFs into two groups which exhibit inverse surface effects that have opposing size-dependence on $a/h$. Further, it can also be seen from figure 3(a) that when $h$ changes from 20 nm to 2 $\mu$m the absolute value of $(V_{cr} - V_{cr0})/V_{cr0}$ decreases considerably and its dependence on $a/h$ becomes very weak. Our numerical study shows that at $h > 720$ nm the absolute value of $(V_{cr} - V_{cr0})/V_{cr0}$ is smaller than 5% throughout the range of $a/h$ considered. In other words, for PNFs of $a/b = 0.25$, the surface effect is negligible when $h$ exceeds this critical value $h_{cr} = 720$ nm. Similar behaviour of the surface effect is observed in figures 3(b) and (c) for the PNFs of ratio $a/b = 0.5$ and 1, respectively. But the critical aspect ratio $(a/h)_{cr}$ increases to 23.1 and 29.4 (figures 3(b) and (c)). In the meantime, our calculation shows that the critical thickness $h_{cr}$ decreases to 580 nm and 300 nm, respectively.

The above size-dependence of $(a/h)_{cr}$ is obtained for constant surface stresses $\sigma_{11}^{(0)} = \sigma_{22}^{(0)} = 1$ N m$^{-1}$ and $a/b = 0.25, 0.5$ or 1. However, $\sigma_{11}^{(0)}$ and $\sigma_{22}^{(0)}$ may vary in the range 0.1 to 1N m$^{-1}$ [27, 28], which could alter the critical value $(a/h)_{cr}$. In addition, the ratio $a/b$ usually changes continuously in the applications of PNFs. It is thus of interest to further study the dependence of $(a/h)_{cr}$ on the residual surface stresses and
yield for PNFs with positive residual surface stresses and (obtained by considering surface effect and residual surface tensions)

theory, i.e. equation (23), can be used. Equations (23) and (26) thin PNFs where (a/h) the ratio a/h can be obtained

based on equation (27) when (a/h) is satisfied, i.e. the surface effect vanishes (a/h)cr to change with the residual surface

There is one obtained without considering surface effect. the ratio a/b. To simplify our analysis we consider relatively thin PNFs where (a/h)cr > 20. In this case, Kirchhoff plate theory, i.e. equation (23), can be used. Equations (23) and (26) yield

\[
\frac{V_{cr}}{V_{cr}^{0}} = \left[ \frac{1}{1 + 2\varepsilon_{51}/(\varepsilon_{31}h)} \right] \cdot \left\{ 1 + \frac{6\pi^2[\tilde{c}_{11}(1 + \beta^2) + 2(\tilde{c}_{12} + 2\tilde{c}_{44})\beta^2]}{\pi^2h[\tilde{c}_{11}(1 + \beta^2) + 2(\tilde{c}_{12} + 2\tilde{c}_{44})\beta^2]} \right\}
\]

(27)

where \(\alpha = a/h\) and \(\beta = a/b\). \((a/h)_{cr} (> 20)\) can be obtained based on equation (27) when \(V_{cr}/V_{cr}^{0} = 1\) is satisfied, i.e. the surface effect vanishes

\[
\left( \frac{a}{h} \right)_{cr} = \left[ \frac{[\pi^2\varepsilon_{51}[\tilde{c}_{11}(1 + \beta^2) + 2(\tilde{c}_{12} + 2\tilde{c}_{44})\beta^2] - 3\pi^2\varepsilon_{31}[\tilde{c}_{11}(1 + \beta^2) + 2(\tilde{c}_{12} + 2\tilde{c}_{44})\beta^2]}{12\varepsilon_{31}\sigma_{11}^{0}(1 + \beta^2)} \right]^{-1/2}.
\]

(28)

Based on equation (28) we show graphically in figure 4 the tendency of \((a/h)_{cr}\) to change with the residual surface (tensile) stresses and the ratio a/b. This figure provides guidance for evaluating the surface effect and its size-dependence on rectangular PNFs with residual surface tension.

Subsequently, we shall consider PNFs where negative residual surface stresses \(\sigma_{11}^{0} = \sigma_{22}^{0} = -1 \text{ N m}^{-1}\) are found. The relative change in the critical buckling voltage \((V_{cr} - V_{cr}^{0})/V_{cr}^{0}\) is calculated in figure 5 against a/h for the PNFs whose thickness h is 20 nm, 200 nm and 2 \(\mu\)m, respectively, and ratio a/b equals (a) 0.25, (b) 0.5 and (c) 1. In this case, negative \((V_{cr} - V_{cr}^{0})/V_{cr}^{0}\) is obtained throughout the range of the aspect ratio a/h considered. Its absolute value increases monotonically with rising a/h. These results indicate that with residual surface compression the surface layers always reduce the critical buckling voltage \(V_{cr}\) and the reduction of \(V_{cr}\) becomes greater for thinner PNFs of a greater aspect ratio a/h.

This behaviour is qualitatively different from that observed in figure 3 for the PNFs with residual surface tension. On the other hand, similar to what is shown in figure 3, the surface effect measured by the absolute value of \((V_{cr} - V_{cr}^{0})/V_{cr}^{0}\) decreases substantially when h increases from 20 nm to 200 nm and to 2 \(\mu\)m. In particular, it is seen in figure 5 that when PNFs are sufficiently thin, e.g., h = 20 nm and a/h = 34.5 in figure 5(a), \((V_{cr} - V_{cr}^{0})/V_{cr}^{0}\) is equal to negative one, i.e. \(V_{cr} = 0\). This suggests that the intrinsic buckling would occur for thin PNFs due to the residual surface compression without any external mechanical or electrical loads. The experimental observation of this phenomenon has not yet been reported for PNFs in the literature. But the theory may provide a physical
The relative change in the critical buckling voltage \((V_{cr} - V_{cr}^0)/V_{cr}^0\) as a function of the length-to-thickness ratio \(a/h\) obtained for PNFs with negative residual surface stresses and (a) \(a/b = 0.25\), (b) \(a/b = 0.5\), (c) \(a/b = 1\).

3.3. Physical mechanisms of the surface effect

In principle, the surface effect is the resultant effect of the surface piezoelectricity, residual surface stress and surface elasticity. To understand the surface effects obtained in section 3.2 we shall first estimate the contributions of the three individual factors to the resultant effect and then study the physical mechanisms via which they exert influence on the buckling of PNFs.

First let us consider PNFs with residual surface tension \(\sigma_{ss}^0\) and residual surface stress \(\sigma_{cs}^0\). In figure 6 \((V_{cr}^0 - V_{cr}^0)/V_{cr}^0\) and \(V_{cr}^0 - V_{cr}^0)/V_{cr}^0\) are obtained for the PNFs under the conditions \(\sigma_{ijkl}^0 = 0\), \(c_{ijkl}^0 = 0\), \(e_{ijil}^0 \neq 0\), \(\sigma_{ijkl}^0 \neq 0\), \(c_{ijkl}^0 = 0\), \(e_{ijil}^0 \neq 0\), \(\sigma_{ijkl}^0 \neq 0\), \(c_{ijkl}^0 \neq 0\), \(e_{ijil}^0 \neq 0\), respectively. These quantities measure the surface effect on the critical buckling voltage due to the surface piezoelectricity, residual surface stress, surface elasticity and all the three factors (i.e. the resultant surface effect). Here the film thickness is fixed at \(h = 20\) nm, the aspect ratio \(a/h\) increases from 5 to 50 and the ratio \(a/b\) is equal to 0.5 (figure 6(a)) or 1 (figure 6(b)). First we see from figure 6 that the effect of the surface elasticity is negligible as compared with that of the surface piezoelectricity and residual surface stresses. This can be attributed to the fact that (1) the surface elasticity-induced increase in the bending stiffness \(D_{ijkl}\) (equation (16)) is too small to significantly change the critical buckling voltage. (2) Its contribution to the in-plane stresses (equation (13)) via the in-plane extensional stiffness \(K_{ijkl}\) (equation (17)) is completely neglected as \(\gamma_{ij}\) is assumed to be zero in this study. Thus in the following discussion we shall only consider the effect of the residual surface stresses and surface piezoelectricity.

In figure 6 the inverse effects on the critical buckling voltage are achieved for the residual surface tension and surface piezoelectricity, i.e. the former enhances the critical buckling voltage while the latter lowers it down. Moreover, strong size-dependence is observed for the effect of the surface tension (equation (16)) of the surface piezoelectricity (equation (17)) described in section 3.2 (also see figure 2). As noted in figure 6, at \(a/h < (a/h)_{cr}\), the effect of the surface piezoelectricity is stronger, which yields negative \((V_{cr}^0 - V_{cr}^0)/V_{cr}^0\). The effect of the surface tension, however, rises with growing \(a/h\). When \(a/h\) approaches \((a/h)_{cr}\) it becomes strong enough to counteract the effect of the surface piezoelectricity and results in \((V_{cr} - V_{cr}^0)/V_{cr}^0 = 0\). Subsequently, at \(a/h > (a/h)_{cr}\) the effect of the surface tension becomes greater and leads to positive \((V_{cr} - V_{cr}^0)/V_{cr}^0\). Similar calculation is also performed for the PNFs with residual surface compression \(\sigma_{cs}^0 = 0\), \(\gamma_{ij}^0 = -1\) N/m². The results are presented in figure 7 where both the surface compression and the surface piezoelectricity decrease the critical buckling voltage. The size-dependences of \((V_{cr}^0 - V_{cr}^0)/V_{cr}^0\) and \((V_{cr}^0 - V_{cr}^0)/V_{cr}^0\) in figure 7 are found to be analogous to their counterparts in figure 6. As a result, \((V_{cr} - V_{cr}^0)/V_{cr}^0\) representing the resultant surface effect is always negative and its magnitude increases monotonically with the rising \(a/h\) (figures 5 and 7).

It is noticed in equation (13) that the significant effect of the residual surface stresses obtained in figures 2, 5, 6 and 7 originates solely from the in-plane stresses \(2\sigma_{11}^0\) and \(2\sigma_{22}^0\).
due to the existence of the top and bottom surface layers. In other words the residual surface stresses change the critical buckling voltage by altering the effective structural rigidity of PNFs. Here the constant surface stresses are considered, whose influence on the buckling naturally increases when less rigid PNFs of greater $a/h$ and (or) smaller $a/b$ are concerned. This explains the size-dependence achieved in figures 6 and 7 for the effect of the constant residual surface stresses.

In addition, equations (13) and (16) show that piezoelectricity affects the structural responses of PNFs by (1) changing the equivalent bending stiffness via the electromechanical coupling, i.e. $(e_{ij3}/k_{33}) \cdot (e_{ij3} \cdot I + (h^2/2) \cdot e_{ij3})$ (equation (16)) and (2) adding the equivalent in-plane stresses through the inverse piezoelectric effect, i.e. $[e_{ij3} + (2/h) \cdot e_{ij3}] \cdot V$ (equation (13)). In particular, the latter is actually the driving force that initiates the buckling of a PNF subject to an electrical voltage $V$. For the PNFs considered the change of the bending stiffness $(e_{ij3}/k_{33}) \cdot [e_{ij3} \cdot I + (h^2/2) \cdot e_{ij3}]$ is from around 3% to 6% of $c_{ijkl}$ at $1$ when $h$ varies between 20 and 200 nm. Here $c_{ijkl}$ is the bending stiffness of the elastic counterparts of the PNFs. Our numerical study shows that the effect on $V_{cr}$ due to such a small change in the bending stiffness can be neglected in the buckling analysis. Thus the piezoelectricity (including the surface piezoelectricity) influences the structural responses of PNFs primarily through the equivalent in-plane stresses $[e_{ij3} + (2/h) \cdot e_{ij3}] \cdot V$ that are proportional to the applied voltage $V$ and the equivalent piezoelectric coefficient $(e_{ij3} + (2/h) \cdot e_{ij3})$. Based on these analyses we conclude that the surface piezoelectricity alters the critical buckling voltage as it generates extra in-plane stresses $(2/h) \cdot e_{ij3}^s \cdot V$ on the PNFs by raising the equivalent piezoelectric coefficient from $e_{ij3}$ of bulk material to $e_{ij3} + (2/h) \cdot e_{ij3}^s$ at the nanoscale. Moreover, in equation (27), it can be shown that

$$6\pi^2[e_{i1}^p(1 + \beta^2) + 2(\bar{c}_{ij2} + 2\bar{c}_{ik4})\beta^2] + 24\alpha^2\sigma_{ij}^0(1 + \beta^2)$$

is satisfied under the condition $\sigma_{ij}^0 = 0$ and $e_{i1k4} = 0$. Based on this inequality and equation (27) the relative change in the critical buckling voltage due to the surface piezoelectricity can be estimated as

$$\left| V_{cr} - V_{cr}^0 \right| \approx \frac{1}{(e_{ij}/\Delta e) + 1} \quad \text{and} \quad \Delta e = \frac{2}{h} \cdot e_{ij3}^s$$

(29)

which is independent of $a = a/h$ and $b = a/b$ but increases with rising $\Delta e$. Equation (27) and thus equation (29) hold true for thin PNFs of $a = a/h \geqslant 20$ as they are based on Kirchhoff plate theory. However, our numerical results given by Mindlin plate theory show (figures 6 and 7) that equation (29) is still valid even at $5 \leqslant a/h \leqslant 20$. We therefore propose that equation (29) may be used to estimate the effect of the surface piezoelectricity in general cases and theoretically explain its negligible size-dependence on $a = a/h$ and $b = a/b$.

4. Conclusions

A sandwich-plate model is developed for the voltage-induced buckling of piezoelectric nanofilms based on Mindlin’s assumption and the theory of the surface effect. Using the model a detailed investigation is conducted regarding the surface effect on the critical buckling voltage and its dependence on the geometric size of the nanofilms. The major conclusions drawn in this study are summarized as follows:

1. The physical origin of the surface effect is found to be the in-plane stresses produced by (a) the residual surface stresses $\sigma_{ij}^0 = \sigma_{ij}^{02}$, and (b) the increase in the piezoelectric coefficient $(2/h) \cdot e_{ij3}^s$ due to the surface piezoelectricity. The surface elasticity, however, has negligible effect on the critical buckling voltage.

2. The residual surface tension (or compression) increases (or decreases) the critical buckling voltage. The magnitude of the change rises with (a) the growing length-to-thickness ratio $a/h$ and (b) the decreasing length-to-width ratio $a/b$. Differently the surface piezoelectricity here always reduces the critical buckling voltage. The reduction increases with $(2/h) \cdot e_{ij3}^s$ but is (almost) independent of $a/h$ and $a/b$.

3. The influence of the surface residual stresses and surface piezoelectricity yields the following resultant surface effect on the buckling of piezoelectric nanofilms. (a) For the nanofilms with residual surface tension, the surface layers decreases the critical buckling voltage when $a/h$ is smaller than a certain critical value $(a/h)_{cr}$. Such an effect decreases with increasing $a/h$. Nevertheless, the surface effect and its dependence on $a/h$ will be revised immediately once $a/h$ exceeds $(a/h)_{cr}$. (b) For the nanofilms with residual surface compression, the surface layers always decrease the critical buckling voltage. This effect turns out to be stronger for a thinner nanofilms of a greater $a/h$. In particular, due to the surface compression, intrinsic buckling could occur for a thin nanofilm without any external loads.
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