A Self-Limiting Material Growth Triggered and Tunable by Force Through Piezocharge-induced Mineralization

Journal:	*Materials Horizons*
Manuscript ID	MH-COM-04-2024-000498.R1
Article Type:	Communication
Date Submitted by the Author:	24-Jun-2024
Complete List of Authors:	Kitchen, Grant; Johns Hopkins University Sun, Bohan ; Johns Hopkins University Omar , Mostafa ; Johns Hopkins University Eisape, Adebayo ; Johns Hopkins University Kang, Sung; Johns Hopkins University,

COMMUNICATION

A Self-Limiting Material Growth Triggered and Tunable by Force Through Piezocharge-induced Mineralization

Grant Kitchen,[ad] Bohan Sun,[bd] Mostafa Omar,[bd] Adebayo Eisape,[cd] and Sung Hoon Kang*[bd]

[a] Department of Materials Science and Engineering, Johns Hopkins University, Baltimore, 21218, USA
[b] Department of Mechanical Engineering, Johns Hopkins University, Baltimore, 21218, USA
[c] Department of Electrical and Computer Engineering, Johns Hopkins University, Baltimore, 21218, USA
[d] Hopkins Extreme Materials Institute, Johns Hopkins University, Baltimore, 21218, USA
Address correspondence to Sung Hoon Kang, shkang@jhu.edu

Electronic Supplementary Information (ESI) available: Supplementary Notes 1-8, Table S1-S5, Fig. S1-S10. To make it easy for readers to follow the content, the supplementary table and figures have been grouped with the relevant supplementary notes. See DOI: 10.1039/x0xx00000x

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

New Concepts

In this manuscript, we report a novel self-limiting growth mechanism that is controllable by force. In contrast with mechanisms that rely on controlling precursors or monitoring material growth, using force as a tunable parameter allows for the growth profile to be adjusted without sophisticated control and monitoring equipment. In addition, force controlled growth avoids disturbing parameters such as temperature and reagent concentration that can significantly impact material morphology and quality. The behavior and growth of the self-limiting mechanism is analytically derived and tested by forming mineral layers using a piezoelectric substrate in an electrolyte solution under cyclic loading conditions to verify that mineral growth decreases the driving force for further growth and that the thickness of the resulting minerals can be accurately predicted based on loading conditions. The findings contribute to the fundamental understanding of self-limiting material growth and can be applied to further material classes and find utility in applications requiring autonomously controlled growth that can adapt to environmental conditions.

Abstract

Controlling the growth of material is crucial in material processing for desired properties. Current approaches often involve sophisticated equipment for controlling precursors and monitoring material formation. Here a self-limiting material growth mechanism controlled by the experienced mechanical loading environment without the need for precise control over precursors or monitoring material growth is reported. Material formation that reduces the driving force for growth is hypothesized to result in a saturation thickness that is dependent on the maximum driving force. Analytical relations based on the growth model are derived and verified using a piezoelectric substrate in an electrolyte solution under cyclic loading at a fixed frequency to attract surrounding mineral ions to form mineral layers. Accumulating mineral layers decrease the driving force for further growth and the material eventually reaches a saturation thickness. This allows for loading force to control the saturation thickness of the self-limiting material growth. Experimental data supports the predicted exponential relations, offering guides to predict the saturation thickness and control the growth profile. The findings are envisioned to contribute to the fundamental understanding of the self-limiting material growth mechanism and could benefit a range of applications including coatings for orthopedic implants as well as marine surface and underwater vehicles.

As the performance of a coating or a device depends on the thickness of a material layer, it is crucial to control the thickness during manufacturing. Current approaches for thickness control tend to require dedicated equipment with control or monitoring systems[1-6]. Control over the final thickness of a fabricated material requires either using approximations to calculate the expected material thickness or continuous measurements to terminate the growth process. For example, a coating can be evaporated to a target thickness that can be estimated based on the expected deposition rate or approximated by measurements with a quartz crystal exposed to the same evaporation conditions[2]. Alternatively, a coating thickness can be estimated and controlled by spin rate and time in spin-coating or by the number of dipping cycles in layer-by-layer growth[7,8].

As an alternative to conventional fabrication and control methods, self-limiting material growth can provide a simple way to control the final thickness of a material. With a self-limiting growth process, additional control or measurements are not needed to accurately terminate the growth process. Instead, input parameters can be modulated to determine the steady-state thickness without a risk of overgrowth.

Current examples of self-limiting material growth include self-assembling monolayers (SAMs), atomic layer deposition (ALD), and passivation layer formation. SAMs are molecular assemblies formed by self-terminating chemical reactions between reactants and substrates to form monolayer-thick coatings that impart desired surface or electrical properties[9,10]. Building on SAMs, post-transition metal chalcogenide compounds (PMTC) can be formed to create bilayers on existing monolayers[9-13]. ALD is a deposition technique that uses sequential gas-phase precursors that react with the surface to form a monolayer. The process reaches saturation due to limitations from the number of reactive surface sites and the steric hindrance of precursor ligands. Oxide passivation layer growth can be self-limiting for substrates such as copper[3], silicon[4,6,14,15], and tungsten[4]. Copper oxide formation starts rapidly due to fast surface reactions, but after island coalescence further oxide growth requires the slower bulk diffusion of copper which decreasing the growth rate as the oxide thickness increases[3].

One of the most significant challenges of current self-limiting material growth is the difficulty in tuning the final thickness. SAMs, liquid metal surface oxides, and two-dimensional PMTCs can form only mono- or bi-layer films, which are unsuitable for thicker layers[11-13]. ALD and other layer-by-layer techniques need precise control over the number of alternating cycles which require specialized equipment and face a tight viable temperature range[1,16]. Oxide passivation layer growth has a limited adjustable range for temperature and oxygen content, restricting the tunability of the growth rate and thickness[6,9,10,14].

To address the challenges, we hypothesized that if the driving force for material growth is decreased by the formation of a material layer, the material will ultimately stop growing at a terminal thickness as the driving force converges to zero. For such a mechanism to be self-limiting, the forming layers must have material properties that counter the driving force. For example, in a material formed by coulombic interactions, the material must be an insulator to diminish the electrostatic force, and the material must be formed through ionic bonds that are drawn through coulombic interaction, thus reducing further material growth. Over time the force of the coulombic interaction is insufficient to attract new material for further growth, resulting in the cessation of growth as in (Fig. 1a). Moreover, the terminal thickness will be dependent on the magnitude of the driving force, as more material will be needed to decrease a larger driving force. These requirements set the range of suitable materials to be ionic compounds, such as minerals. In maritime applications, suitable minerals that could be formed from ionic components seawater include carbonates such as calcite, aragonite, vaterite, and amorphous calcium phosphate, sulfates such as gypsum and celestite., and organic crystals including calcium tartrate and calcium malate. In the body, the ion constituents of bodily fluids could make it suitable for the formation of phosphates such as apatite, brushite, dahllite, and amorphous calcium phosphate as well as similar organic crystals that are possible to form in seawater[17]. The ionic composition of seawater, blood plasma, and simulated body fluid utilized in the study are listed in Table S3, and feasible compounds to form in these fluids are shown in Table S4. However, the constitution of the surrounding fluid is not limited by natural availability, as the addition of components such as iron, barium, zinc, silicon, and copper to the surrounding solution could allow for a wider range of ionic compounds to form, including sulfides, chlorides, oxides, perovskites, arsenates, fluorides, hydroxides, and organic crystals in addition to otherwise inaccessible phases of phosphates, sulfates, and organic crystals as shown in Table S5.

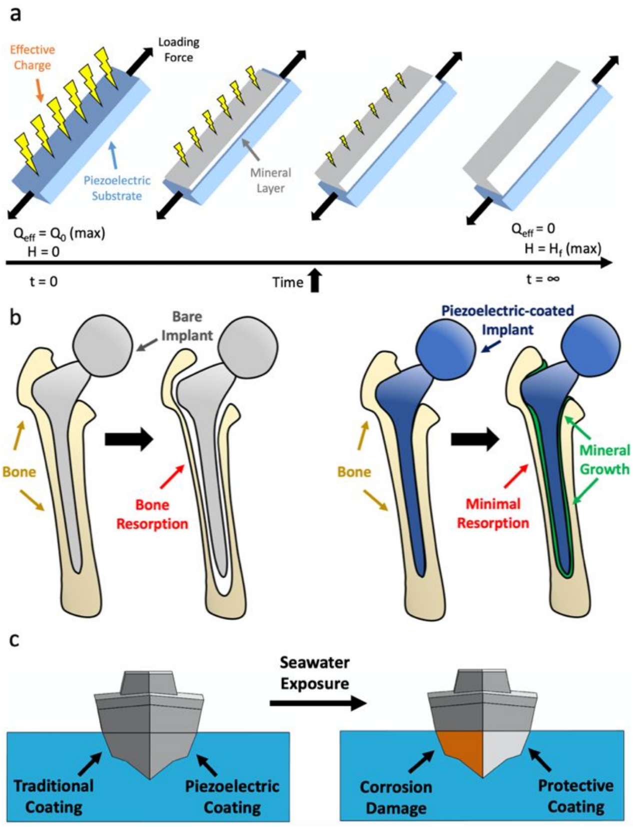


Fig. 1 Schematic of the proposed mechanism and potential application areas. (a) Self-limiting material growth model; the mechanical deformation of a piezoelectric substrate generates surface charges Q_0 that attract mineral ions and lead to formation of mineral layers which decrease the effective charge Q_{eff}, eventually reaching a saturation thickness H_f as the effective charge converges to zero. (b) Self-limiting material growth utilized to limit stress shielding and bone resorption from an orthopedic implant to improve the long-term stability of the implant. (c) Self-limiting material growth utilized to create a regenerative protective coating for maritime vessels to protect against corrosion caused by extended exposure to seawater.

Such a mechanism has several advantageous properties for a range of applications. For example, orthopedic implants need to promote ingrowth with the surrounding tissue, but uncontrolled growth can damage the surrounding tissue and lead to stress shielding and bone resorption due to mismatch between the stiffness of the implant and surrounding tissue that cannot be attenuated due to the inflexibility of the implant to change its mechanical properties over time[18,19]. A self-limiting growth mechanism could instead promote beneficial mineralization without the risk of overgrowth and provide a means for continued regeneration as in (Fig. 1b). Under changing loading demands, the self-adaptive implant could regulate both the modulus of the material and the ingrowth of minerals into the surrounding tissue, allowing the implant to minimize tissue resorption and maintain interface stability with tissues over time. Additionally, maritime vessels experience corrosion damage from prolonged exposure to seawater. Traditional protective methods include periodically applying coatings and cleaning the vessels. Instead, a self-limiting growth mechanism could form a protective surface layer that can regenerate over time without risk of overgrowth that can create drag and friction as in (Fig. 1c).

To verify the hypothesis, we considered a piezoelectric substrate cyclically stressed while submerged in an electrolyte solution to generate piezoelectric charges that attract surrounding mineral ions to adsorb and form mineral layers[20]. As electrically insulating mineral layers form, the force due to the underlying charges become progressively shielded, decreasing the driving force for further growth in a self-limiting manner. If we consider the piezoelectric charge magnitude as a constant value, then we can consider an effective charge as the charge acting on the surrounding mineral ions. Then, we can write Equation (1) and (2) based on the proposed model and derive solutions as Equation (3) and (4). (See Supplementary Note 1 for details.)

$$\frac{dQ_{eff}}{dt} = -\alpha Q_{eff} \qquad (1)$$

(Q_{eff}: effective charge, t: time, α: proportionality constant)

where $Q_{eff} = Q_o$ at $t = 0$ and $Q_{eff} = 0$ at $t = \infty$.

$$\frac{dH}{dt} = -\beta H \qquad (2)$$

(H: thickness, t: time, β: proportionality constant)

where $H = 0$ at $t = 0$ and $H = H_f$ at $t = \infty$.

Then, the solutions of Equation (1) & (2) are Equation (3) & (4):

$$Q_{eff}(t) = Q_0 \exp(-\alpha t) \qquad (3)$$

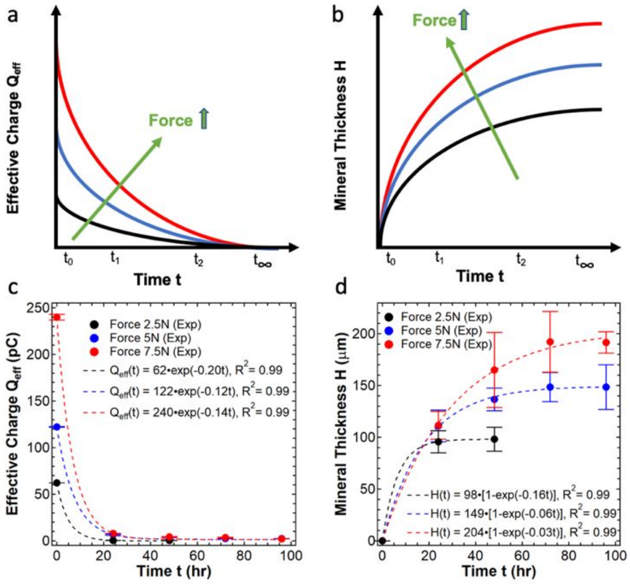
(Q_0: initial charge)

$$H(t) = H_f[1 - exp(-\beta t)] \qquad (4)$$

(H_f: final thickness)

Initially, the generated charges are at a maximum with no material (mineral in this example) formed. As materials form, the generated charges are separated by the insulating materials, decreasing the force experienced by the surrounding ions, therefore decreasing the driving force for further growth. At infinite time, there is no effective charge and the mineral thickness reaches a saturation thickness. The analytical models indicate that the effective charge decays exponentially over time, and the maximum thickness is determined by the initial charge magnitude (the unshielded charge magnitude), which is determined by the applied force and the piezoelectric coefficient. In this system, force can be used as a parameter to control self-limiting growth. By utilizing force as a tuning parameter, growth could be adjusted without requiring changes in growth environment (e.g. temperature or substrate geometry[21]) and sophisticated equipment. Furthermore, with the final thickness controlled by force, the mechanism could work for a wide range of thicknesses and the force response can be further adjusted by changing the piezoelectric coefficient of the substrate.

We conducted experiments to verify our model by measuring the effective charge on the mineral surface and mineral thickness over time (See Supplementary Notes 2-5 for details). We further verified our hypothesis and model by studying the effects of varying cyclic loading force amplitudes. If the force magnitude is increased, the magnitude of the generated charge increases as the charge generation from a piezoelectric material is proportional to the applied force. These expected relations under fixed loading frequency conditions are shown in (Fig. 2a) and (Fig. 2b). In all cases, under a consistent loading the minerals reach a state where the driving force is insufficient for further growth, leading to a saturation thickness.



$Q_{eff}(t) = Q_0 \exp(-\alpha t)$ $\quad$ $H(t) = H_f[1 - \exp(-\beta t)]$

Fig. 2 Schematic and experimental data of the effective charge and the mineral thickness as a function of time. (a) Expected behavior of effective charge as a function of time at different force amplitudes. (b) Expected behavior of mineral thickness as a function of time at different force amplitudes. (c) Measured effective charge as a function of time with different force amplitudes at a fixed frequency of 2 Hz and fitting results, with points and error bars representing the average and standard deviation of 12 measured charge values. (d) Measured mineral thickness as a function of time with different force amplitudes at a fixed frequency of 2 Hz and fitting results, with points and error bars representing the average and standard deviation of 60 measured thickness values.

Experimental data showed good agreements with the analytical solution as (Fig. 2c) and (Fig. 2d), with exponential relations between charge vs. time and thickness vs. time. From the data analysis, the proportionality constant α in Equation (1) and (3) had a value of (0.15 ± 0.03) while the proportionality constant β in Equation (2) and (4) decreased with the increasing force. The initial charge Q_o showed a good agreement with the estimation based on an applied force and a piezoelectric coefficient at a fixed frequency of 2 Hz as in (Fig. 3a) and in Equation (5):

$$Q_o \sim \text{force} \bullet \text{piezoelectric coefficient} \qquad (5)$$

where the piezoelectric coefficient of the PVDF depends on the loading frequency as it can affect piezoelectric properties of the PVDF[22-26]. Therefore, we can choose a force value at a given loading frequency to have the desired final thickness. From the data analysis, mineral growth stopped at an effective charge magnitude Q_o of (1.52 ± 0.89 pC). Moreover, higher force amplitudes resulted in higher initial charges and final thicknesses while converging to similar effective charge amplitudes at the saturation thicknesses.

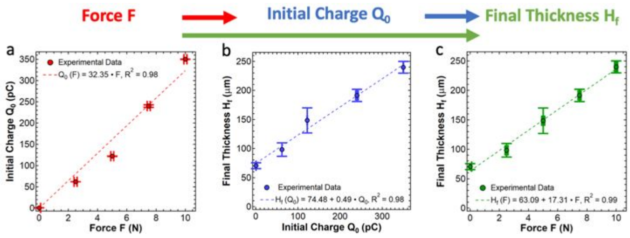


Fig. 3 Relations between force, initial charge, and final thickness. (a) Measured initial charge as a function of force amplitude and fitting results. (b) Measured final thickness as a function of initial charge and fitting results. (c) Measured final thickness as a function of force amplitude and fitting results. The points and error bars represent the average and standard deviation of each measured value. For all values, a fixed frequency of 2 Hz was used; For charge values, 12 measurements are used; For thickness values, 60 measurements are used; For force values, 200 measurements are used.

Based on the experimental data, we analyzed the relation between the initial charge and the final thickness as (Fig. 3b), which showed almost a linear relation within our measured data range. The results show that using force as a parameter to control the final thickness is a viable way to have self-limiting material growth as (Fig. 3c). Additionally, we have found strong correlations between the piezoelectric coefficient of the material, applied force, and initial charge that can be related to the final thickness. The main differentiator of our mechanism compared to existing self-limiting growth mechanisms is that the self-limiting behavior relies on the gradually decreasing driving force which can be modulated by the applied force. The mechanism is not dependent on changing temperature or reagent concentration to alter the growth, and it does not rely on specific chemical bonding or molecular design. In addition, further growth after saturation is possible if progressively stronger force is provided, while a fixed cyclic loading amplitude will reach a saturation thickness.

The temperature dependence of the self-limiting growth mechanism was studied by mineralizing unloaded samples until steady state mineral thickness was reached at a temperature range of 25–50°C (See Supplementary Note S7 for details). In this case, the driving force for mineralization was due to the inherent charges present on the piezoelectric substrates. In these experiments, there was no statistically significant difference in the ultimate film thickness as a function of temperature, nor was there a statistically significant difference in the mineral thickness between samples mineralized at the same temperature. The measurement results are summarized in Table S2. This indicates that the ultimate mineral thickness is dependent on and controllable by force, and further validates our model indicating that the charge magnitude is the primary driving force and is reduced predominantly by distance from the charge source. In addition, previous studies indicated that the initial film growth rate was strongly correlated with temperature over the initial 24 hours of film growth. This could indicate that the speed of the film growth can be accelerated with increasing temperature, but the mineral thickness will eventually converge to the same ultimate thickness. Previously, we reported the material characterization data of the minerals grown on piezoelectric scaffold[27]. In that study, we found that when grown at a temperature of 25°C, the minerals had a modulus of 0.75 ± 0.58 GPa, a hardness of 6.25 ± 5.59 MPa as measured by nanoindentation, and predominantly displayed hydroxyapatite peaks in x-ray diffraction (XRD) analysis. In comparison, when grown at a temperature of 50°C, nanoindentation measurements determined the minerals to have a modulus of 7.37 ± 11.87 GPa and a hardness of 0.73 ± 2.81 GPa. This indicates that while temperature does not affect the ultimate mineral thickness, it can impact the crystallinity and elastic properties of the formed mineral layers.

We envision that our mechanism opens opportunities to investigate and design other material systems and synthesis processes with self-limiting growth behaviors. The particular example that we reported in this paper can be beneficial for orthopedic implants to minimize overgrowth, stress shielding, and bone resorption, improving the long term stability of implants. By allowing for the mechanical properties and connecting interface of the implant to adapt to meet loading demands and changes in the underlying tissues, the implant could improve long-term outcomes by reducing mechanical property mismatches and maintaining solid contact with the anchored tissue. In particular, our system can repeatedly regenerate over time, utilizes solutions with the same ionic constituents as blood, and forms biocompatible calcium phosphate similar to bone tissue[9,10,18,19,28]. In addition, some piezoelectric materials such as ZnO have been found to prevent biofilm formation, which could also provide anti-biofouling properties to the surface of the implants that can limit degradation over time[28]. While we used a mechanical loading machine for stimulating the piezoelectric matrix, one can also use other sources of mechanical energy such as ultrasound or vibration to generate charges[29-31]. The speed of the material growth can be further accelerated by increasing the SBF temperature and concentration. Such a self-limiting growth mechanism could be also beneficial for creating regenerative smart coatings to provide anti-abrasion or anti-corrosion properties to surfaces while maintaining controlled thicknesses to limit unwanted friction or drag[9,10]. This could be beneficial for maritime applications with long-term exposure to corrosive environments such as ships and submarines. Typically, a protective paint or coating is applied but over time the material degrades. In a self-limiting growth case, the protective coating could form from surrounding ions in response to shear stress to autonomously protect against corrosion and damage. Furthermore, a force-controlled response mechanism can allow for a material to be mechanically adaptive to external stimuli, which can potentially benefit emerging technologies such as soft robotics and morphological computation[32].

Conclusions

In summary, we report a self-limiting material growth triggered and tunable by force. We hypothesized that if the formation of a material layer decreases the driving force for further growth, the material will stop growing at a saturation thickness as the driving force converges to zero. Based on the hypothesis, we derived the analytical relations between charge vs. time and thickness vs. time. We tested the analytical model by considering a piezoelectric charge-induced mineral growth, where the applied cyclic force amplitude determines the generated piezoelectric charge magnitude that drives mineralization. The experimental results showed good agreements with the analytical model. Furthermore, as higher force amplitude results in a higher charge magnitude, higher force also results in higher saturation thickness. Thus, one can control the final thickness of the self-limiting material growth based on the applied force amplitude and the piezoelectric coefficient of the substrate without sophisticated control and monitoring equipment[9,10]. Such a mechanism could be utilized to enhance the durability and resilience of materials by preventing or slowing degradations and allowing materials to be responsive to changing environmental conditions[32].

Author Contributions

S.H.K. conceived the concept. S.H.K. derived analytical relations. S.H.K. and G.K. designed experiments. G.K. prepared samples and performed experiments. G.K. and S.H.K. analyzed experimental data. G.K., B.S., M.O., A.E., and S.H.K. interpreted data and refined the self-limiting growth model. G.K. and S.H.K. wrote the manuscript. All authors edited the manuscript.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was supported by the Air Force Office of Scientific Research Grants (Award numbers: FA9550-18-1-0073 and FA9550-21-1-0368, Program manager: Dr. Byung-Lip (Les) Lee), Johns Hopkins University Whiting School of Engineering Start-Up Fund and Hanwha Non-Tenured Faculty Award. Any opinions, finding, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the United States Air Force.

References

1 J. Lu, J. W. Elam, and P. C. Stair, *Surf. Sci. Rep.,* 2016, **71,** 410-472.
2 K. L. Choy, *Prog Mater. Sci.,* 2003, **48**, 57-170.
3 J.C. Yang, B. Kolasa, J.M. Gibson, and M. Yeadon, *Appl. Phys. Lett.,* 1998, **73**, 2841-2843.
4 M. Liu, P. Jin, Z. Xu, D.A.H. Hanaor, Y. Gan, and C.Q. Chen, *Theor. Appl. Mech. Lett.*, 2016, **6**, 195-199.
5 F.J. Ma, S.C. Rustagi, G.S. Samudra, H. Zhao, N. Singh, G.Q. Lo, and D.L. Kwong, *IEEE Electr. Device Lett.,* 2010, **31**(7), 719-721.
6 J.T. Wright, D.J. Carbaugh, M.E. Haggerty, A.L. Richard, D.C. Ingram, S. Kaya, W.M. Jadwisienczak, and F. Rahman, *Semicond. Sci. Technol.,* 2016, **31**(105007), 1-10.
7 Y.Y. Lee, G.M. Choi, S.M. Lim, J.Y. Cho, I.S. Choi, K.T. Nam, and Y.C. Joo, *Sci. Rep.,* 2016, **6**(25332), 1-9.
8 H.J. Choi, K.C. Park, H. Lee, T. Crouzier, M.F. Rubner, R.E. Cohen, G. Barbastathis, and G.H. McKinley, *ACS Appl. Mater. Inter.,* 2017, **9**, 8354-8360.
9 A. Ulman, *Chem. Rev.,* 1996, **96**(4), 1533-1554.
10 F. Schreiber, *Prog. Surf. Sci.,* 2000, **65**, 151-256.
11 B.J. Carey, J.Z. Ou, R.M. Clark, K.J. Berean, A. Zavabeti, A.S.R. Chesman, S.P. Russo, D.W.M. Lau, Z.Q. Xu, Q. Bao, O. Kevehei, B.C. Gibson, M.D. Dickey, R.B. Kaner, T. Daeneke, and K. Kalantar-Zadeh, *Nat. Commun.,* 2017, **8**(14482), 1-9.
12 K. Khoshmanesh, S.Y. Tang, J.Y. Zhu, S. Schaefer, A. Mitchell, K. Kalantar-Zadeh, and M.D. Dickey, *Lab Chip,* 2017, **17**, 974-993.
13 K. Kalantar-Zadeh, J. Tang, T. Daeneke, A.P. O'Mullane, L.A. Stewart, J. Liu, C. Majidi, R.S. Ruoff, P.S. Weiss, and M.D. Dickey, *ACS Nano,* 2019, **13**, 7388-7395.
14 X. Zhao, C.M. Wei, L. Yang, and M.Y. Chou, *Phys. Rev. Lett.,* 2004, **92**(23), 236805-1-236805-4.
15 K.Q. Peng, X. Wang, L. Li, Y. Hu, and S.T. Lee, *Nano Today,* 2013, **8**, 75-97.
16 R. Xu, M. Hua, S. Wu, S. Ma, Y. Zhang, L. Zhang, B. Yu, M. Cai, X. He, and F. Zhou, *Matter,* 2022, **5**, 634-653.
17 S. Weiner and P. M. Dove, *Rev. Mineral. Geochem.*, 2003, **54**(1), 1-29.
18 M.L. Raffa, V.H. Nguyen, P. Hernigou, C.H. Flouzat-Lachaniette, and G. Haiat, *J. Orthop. Res.,* 2021, **39**, 1174-1183.
19 T. Wu, S. Yu, D. Chen, and Y. Wang, *Mater.*, 2017, **10**(1187), 1-14.
20 S. Orrego, Z. Chen, U. Krekora, D. Hou, S.Y. Jeon, M. Pittman, C. Montoya, Y. Chen, and S.H. Kang, *Adv. Mater.,* 2020, **32**(1906970), 1-6.
21 G. Biasiol and E. Kapon, *J. Cryst. Growth,* 1999, ***201/202***, 62-66.
22 G.M. Sessler, *J. Acoust. Soc.,* 1981, **70**(6), 1596-1608.
23 A.M. Vinogradov, V.H. Schmidt, G.F. Tuthill, and G.W. Bohannan, *Mech. Mater.,* 2004, **36**, 1007-1016.
24 W. Xia and Z. Zhang, *IET Nanodielectr.,* 2018, **1**(1), 17-31.
25 Q.G. Chi, L. Gao, X. Wang, Y. Chen, J.F. Dong, Y. Cui, and Q.Q. Lei, AIP Adv., 2015, **5**(117103), 1-8.
26 J. Li, Z. Zhu, L. Fang, S. Guo, U. Erturun, Z. Zhu, J. E. West, S. Ghosh, and S. H. Kang, *Nanoscale,* 2017, **9**, 14215-14228.
27 N. Buettner, G. Kitchen, M. Omar, B. Sun, H. Lee, S. H. Kang, and A. T. Akono, *MRS Adv.,* 2023, **8**, 1082-1089.
28 S. Pechook, K. Sudakov, I. Polishchuk, I. Ostrov, V. Zakin, B. Pokroy, and M. Shemesh, *J. Mater. Chem. B,* 2015, **3**, 1371-1378.
29 H. Mohapatra, M. Kleiman, and A.P. Esser-Kahn, *Nat. Chem.,* 2017, **9**, 135-139.
30 Z. Wang, J. Wang, J. Ayarza, T. Steeves, Z. Hu, S. Manna, and A.P. Esser-Kahn, *Nat. Mater.,* 2021, **20**, 869-874.
31 Z. Wang, J. Ayarza, and A.P. Esser-Kahn, *Angew. Chem.,* 2019, **131**, 12151-12154.
32 B.E. Treml, R.N. McKenzie, P. Buskohl, D. Wang, M. Kuhn, L.S. Tan, R.A. Vaia, *Adv. Mater.,* 2018, **30**(1705616), 1-6.

Data availability statements

The data supporting this article have been included as part of the Supplementary Information.