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Authors

Vargo, Emma

Ma, Le

Li, He

et al.

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Functional Composites by Programming Entropy-Driven Nanosheet Growth

Emma Vargo[1,2], Le Ma[1,2], He Li[2,3], Qingteng Zhang[4], Junpyo Kwon[2,5], Xiaochen Tang[6], L. Tovmasyan[1], Jasmine Jan[7], Ana C. Arias[7], Hugo Destaillats[6], Wei-Ren Chen[8], William Victoria Heller[8], Robert O. Ritchie[2,5], Yi Liu[2,3], Ting Xu[1,2,9]*

[1]Department of Materials Science and Engineering, University of California, Berkeley, CA 94720, USA

[2]Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[3]The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[4]Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA

[5]Department of Mechanical Engineering, University of California, Berkeley, CA 94720, USA

[6]Energy Technologies Area, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[7]Department of Electrical Engineering and Computer Sciences, University of California, Berkeley, CA 94720, USA

[8]Spallation Neutron Source, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA

[9]Department of Chemistry, University of California, Berkeley, CA 94720, USA

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Abstract

Nanomaterials must be systemically designed to be technologically viable.[1-5] Driven by optimizing intermolecular interactions, current designs are too rigid to plug in new chemical functionalities and cannot mitigate condition differences during integration.[6,7] Despite extensive optimization of building blocks and treatments, accessing nanostructures with the required feature sizes and chemistries is difficult. Programming their growth across the nano-to-macro hierarchy also remains challenging, if not impossible.[8-13] To address these limitations, we should shift to entropy-driven assemblies to gain design flexibility, as seen in high entropy alloys, and program nanomaterial growth to kinetically match target feature sizes to the system mobility during processing.[14-17] Here, following a micro-then-nano growth sequence in ternary composite blends, we successfully fabricate high-performance barrier materials composed of >200 stacked nanosheets (125 nm in sheet thickness) with defect density < 0.056 μm^{-2} and ~ 98% efficiency in controlling the defect type. Contrary to common perception, polymer chain entanglements are advantageous to realize long-range order, accelerate the fabrication process (<30 minutes), and satisfy specific requirements to advance multi-layered film technology.[3,4,18] The present studies showcase the feasibility, necessity, and unlimited opportunities to transform laboratory nanoscience into nanotechnology through system engineering of self-assembly.