Stability of 2D Hybrid Lead Halide Perovskites: Perspective from Bulk Crystals and Thin Films

Eugenia S. Vasileiadou
Bin Wang, Ioannis Spanopoulos, Ido Hadar, Alexandra Navrotsky, Mercouri G. Kanatzidis
Department of Chemistry, Northwestern University, Evanston, IL 60208, United States
Eugenivasileiadou2022@u.northwestern.edu

Abstract:
Two-dimensional (2D) hybrid organic-inorganic halide perovskites are a promising class of environmentally stable semiconductors, whose inherent structural tunability and technological features provides a vast compositional space to engineer new materials for optoelectronic applications. Within this compositional space, several different homologous series and structure types of 2D perovksites have been developed for their successful fabrication of solar cells, light-emitting diodes and radiation detectors with attractive efficiencies. Although 2D lead iodide perovskites exhibit superior stability over their 3D parent structures, a systematic understanding of their observed stability is missing. Herein, we actuate a comprehensive study on the relative stability of several distinct families of 2D halide perovskites in bulk crystal and film form. [1],[2] Thermochemical evaluation of the representative 2D structure types of Ruddlesden-Popper (RP) and Dion-Jacobson (DJ) perovskites was undertaken based on calorimetric measurements that reveal that the enthalpy of formation for the RP perovskites is negative while for the DJ perovskites is positive (Figure 1). Film stability tests demonstrate consistent observations with the thermochemical findings, where RP lead iodide perovskites are both thermodynamically and environmentally stable candidates for optoelectronic applications. Additionally, the methodical tailoring of the 2D perovskite structure's composition (organic and inorganic component), unveils trends in the comparison of 2D lead iodide and bromide perovskites, for the acquisition of halide perovskites with enhanced stability. Phase-pure synthesis of a new *n*=6 perovskite compound is reported with the characterization of the single crystal structure and optical properties of the material. Structural – crystallographic analysis of bulk, layered perovskites provides atomistic insight to control these materials' stability. Our work highlights the importance of the rational assessment of stability in 2D hybrid halide perovskites for the optimal synthetic design and engineering of environmentally robust perovskite materials for next-generation optoelectronic devices.

REFERENCES

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FIGURES

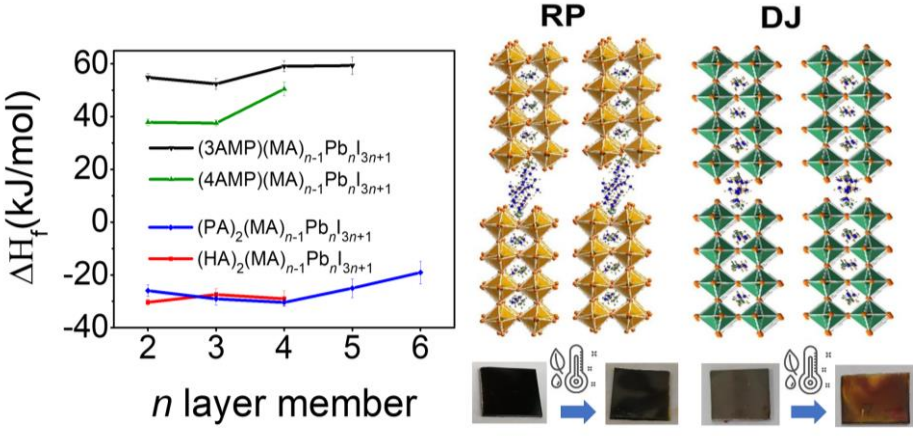

Figure 1: Enthalpy of formation as a function of perovskite layer thickness (*n*) for the four series of RP and DJ perovskites studied, incorporating the spacer cations: pentylammonium (PA), hexylammonium (HA), 3- and 4-(aminomethyl)piperidinium (3AMP) and (4AMP) respectively, alongside schematic of RP and DJ structure with corresponding films.