The development of advanced thin film synthesis techniques over the past several decades has sparked a renaissance in the design of nanomaterials for clean energy and quantum computing technologies. While it is now possible to produce oxide and semiconductor thin films in almost limitless configurations, engineering of desirable functionality for device applications depends on precise control of atomistic structure and defects. Complex synthesis pathways can lead to significant deviations from idealized structures, which occur at length scales that are challenging to probe experimentally and theoretically. This task is further compounded by dynamic changes imparted by processing steps and subsequent exposure to extreme environments. Here I will discuss a materials design strategy based on precision synthesis, theory calculations, and atomistic characterization, grounded in emerging data science tools that enable rich, quantitative analysis at scale. Our results illustrate how the full range of information from modern, data-infused electron microscopy can unlock promising new materials for energy storage, electronics, and computing.