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Thin Film Synthesis of New Complex Titanates.

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Thin film deposition methods allow for one to synthesize rationally specific compositions in targeted crystal structures. Because most of the thermodynamic and kinetic variables that control the range of materials that can be synthesized are unknown for specific compounds/processes, epitaxial stabilization and design of artificially layered crystals are driven through empirical investigations. Using examples taken primarily from the family of complex titanates, which exhibit a range of interesting physicochemical behaviors, the thermodynamic and kinetic factors that control materials design using thin film deposition are discussed. The phase competition between the pyrochlore and the (110) layered perovskite structure in the $RE_2Ti_2O_7$ family ($RE =$ rare-earth, Bi) will be explored, using pulsed laser deposition as a synthesis method. For $RE =$ Gd, Sm, Nd, and La, the phase stability over a wide range of conditions is dictated entirely by substrate choice, indicating that the free energies of the phases are similar enough such that by controlling nucleation one controls the phase formation. In a related fashion, the growth of $AETi_2O_5$ films ($AE =$ Ba or Sr) will be discussed with respect to the formation of single-phase films or films that phase separate into $AETiO_3$ and TiO_2 . The entire $Ba_{1-x}Sr_xTi_2O_5$ series was grown and will be discussed with respect to growth technique (using MBE and PLD) and/or substrate choice. In this case, rock-salt substrates, which are not expected to interact strongly with any phase in the system, allow for the formation of single-phase films. Finally, several examples will be discussed with respect to the $(SrO)_m(TiO_2)_n$ system, which includes the perovskite $SrTiO_3$ and the Ruddlesden-Popper phase Sr_2TiO_4 , grown using layer-by-layer molecular beam epitaxy. The solid phase epitaxial formation of the perovskite $SrTiO_3$ from superlattices of rock-salt SrO and anatase TiO_2 is discussed from both a kinetic and thermodynamic perspective by exploring the growth of a range of m and n values. Using similar arguments for stability, new layered intergrowths in the Sr_mTiO_{2+m} family are presented and their structures are discussed.