Plasma synthesis of nanomaterials offers potentially higher throughput, lower cost, and better control of material properties than conventional chemical methods. Over the past few years we have conducted comprehensive fundamental studies of nanomaterials synthesis by atmospheric pressure arc discharges. This work has led to significant advancements in the understanding of the synergistic roles of plasma and materials processes in the arc synthesis of carbon and boron-nitride nanomaterials. In order to understand nanostructure formation we needed to determine the plasma and gas composition conditions in the nucleation and growth region. This data was not available and well-known before, because it is difficult to measure plasma parameters inside the arc. We determined plasma parameters in the growth region using various in-situ plasma diagnostics and fluid modelling. Additionally, atomistic simulations helped to analyse crucial processes in nanomaterial synthesis. The dominance of diatomic carbon molecules in the arc periphery, a probable pre-cursor species for synthesis of carbon nanostructures, and the dominance of C atoms in the arc core are important new findings of these studies. For boron nitride nanotubes two possible mechanisms of synthesis root-growth from boron clusters and volumetric growth from boron-nitride nanocages are being investigated. Papers are available at nano.pppl.gov