Dye-sensitized solar cell (DSSC) devices are 3rd generation photovoltaic (PV) devices, known for their simplified device construction and delineation of charge generation and transport characteristics. Conventional manufacturing of DSSC devices encompasses doctor blade or spin coat printing technologies for successive deposition of thin film layers; while sufficient, these film-based print technologies highlight constraints regarding sufficiency of ink viscoelasticity and extrudate processivity for microstructure control. Ongoing research regarding DSSC device manufacturing and performance characterization reference the relationship between ink morphology, ink printability, and thin film microstructure variation on photovoltaic efficiency through affecting ionic, or electronic, charge transport efficiencies. This dissertation work positions direct ink writing (DIW), an additive manufacturing (AM) technique, as a tool to further assess the relationship between an ink's morphological properties, rheological properties, and printability, on the extrudate's microstructure and resulting DSSC device performance. Key results demonstrate that modification of 1) ink morphology (polymer concentration [8, 16, 24 wt.%]), 2) DIW extrusion parameters (nozzle diameter [100, 330, 410, 510 μm], translational nozzle velocity [1 – 50 mm/s], nozzle height [50 – 1400 μm], extrusion pressure [5 – 420 PSI]), and 3) extrudate processing conditions (thermal curing, *in situ* UV irradiation) allows for tunability of ink printability and extrudate film microstructure with greater precision than demonstrated thus far utilizing conventional doctor blade and spin coat print technologies. Referencing assembled DSSC devices utilizing liquid electrolytes as reference, addition of polymer material for printability optimization permits conventional assembly of DSSC devices with tunable performance values normalized to 80.7%, 65.1%, and 26.3% efficiency, with respective efficiency variations of -8.7%, -9.4%, and +13.3% when DSSC devices are assembled via DIW hybrid-AM technologies. The magnitude of DSSC efficiency variation was assessed via analysis of variance (ANOVA) statistical methodology, whereby factor variation of electrolyte morphology, semiconducting film microstructure, and manufacturing technology were assessed, identifying statistical significance of all three factors on DSSC device performance. In this report, a rigorous experimental regime outlining the synthesis, rheological characterization, printability, and post-processing of direct ink writeable inks is presented, demonstrating semi-autonomous manufacturing capabilities of DSSC devices utilizing DIW print technologies.