Over 44 million Americans rely on dentures, and the demand is expected to grow with an aging population. Poor denture performance can compromise oral health, which is closely linked to systemic conditions such as cardiovascular disease and diabetes. By developing denture base materials that are both tougher and more adaptable to oral tissues, this research addresses a critical need for more durable, comfortable, and cost-effective prosthetic solutions. These innovations represent a significant step toward next-generation prosthodontics, where advanced materials and digital manufacturing converge to improve patient outcomes and accessibility. The rapid advancement of digital dentistry has allowed for these standards to be reached in a revolutionary manner. However, current denture base materials, primarily polymethyl methacrylate (PMMA), remain limited by brittleness, low fracture toughness, and the need for excessive thickness to ensure mechanical reliability. This study introduces a novel class of urethane-based photopolymer resins designed for multi-material 3D printing, offering improved mechanical performance and adaptability. These resins, formulated with low to ultra-low viscosities and engineered for non-covalent polymer reinforcing interactions, are compatible with both vat photopolymerization and inkjet printing technologies. In particular, the 3D inkjet printing process is an attractive advance that simultaneously delivers multi-materials at high resolution. Experimental formulations were synthesized using a 1:1 molar ratio of urethane to acid functional groups and a 1:1 urethane to monomer ratio. Mechanical testing of photopolymerized specimens that included evaluating flexural strength, modulus, and toughness was conducted under both dry and simulated oral conditions. Several formulations achieved flexural moduli in the target range of 2.5–3.0 GPa, indicating a promising balance between rigidity and flexibility. Higher stiffness denture tooth materials are also needed to achieve greater wear resistance for extended clinical service life. However, water absorption led to reduced performance in wet conditions, prompting further investigation into hydrophobic monomer modifications to enhance resistance to moisture induced degradation.