While cellulose and its derivatives have garnered significant attention for advanced manufacturing purposes, the interest in other bio-derived polymers for similar applications has also been on the rise. One such biopolymer is agarose, a linear polysaccharide composed of repeating agarobiose units extracted from red algae. Much like cellulose, agarose boasts natural abundance, non-toxicity, renewability, and biodegradability, and it possesses unique properties, such as the ability to form thermos-reversible gels and freestanding films. However, both materials share certain limitations, including brittleness, lack of flexibility, and fragility under low moisture conditions. To address these shortcomings, plasticizers have been employed to enhance the flexibility and stretchability of agarose films. Cellulose nanocrystals (CNC), owing to their crystalline nature, have been harnessed as reinforcing agents with the use of CNC with differing polymorphs as supporting agents for plasticized agarose films to exert control over folding behavior. In this study, agarose and cellulose nanocrystals, along with various additives and innovative techniques, will be utilized as promising avenues for developing versatile materials with enhanced mechanical and thermal properties, opening up exciting possibilities for applications such as actuating textiles.