Liquid crystal elastomers (LCEs) are stimuli-responsive materials. Programming the local orientation within LCEs is a promising route to realize 3-D deformations. Recent demonstrations have demonstrated these materials can exhibit exceptional work capacity of upwards of 40 J/kg. However, the inefficient and slow responses of LCEs to heat and light limit the functional integration of these materials in many applications. Electrical control of LCEs has and continues to be the preferred stimuli. Recently, electromechanical actuation of LCEs at ambient conditions was realized by exploiting the unique mechanical anisotropy (e.g., association of modulus with orientation) of aligned LCEs. By coating both sides of an aligned LCE with compliant electrodes and applying voltage, the LCE film is compressed in its thickness, leading to deformation in the planar area. However, due to the mechanical anisotropy, the LCE deforms preferentially in the lower modulus direction, leading to directional mechanical response in these actuating elements. Here, we are concerned with understanding the role of materials properties on the electromechanical response of LCEs. Using a newly developed materials chemistry and employing established processing methods, reliable electromechanical actuation is realized in LCEs with high mechanical anisotropy and uniaxial actuation strains up to 17.5%. The uniaxial actuation behavior is closely predicted using previously developed theory and the physical materials properties of the LCEs. Response times of 1 Hz are demonstrated.