Shape memory wool fabrics having both temperature and moisture responsiveness were fabricated by a functional nanocomposite treatment comprising of shape memory polyurethane (SMPU) and cellulose nanowhisker (CNW). Water vapor permeability and sweat absorption properties were investigated under different temperature or relative humidity values to test smart comfort capabilities of the treated fabrics. Besides, felting shrinkage and weight loss of the fabrics after repeated washing cycles were investigated for end use performance. It was found out that the wool fabrics functionalized by nanocomposite treatments exhibited dynamic breathability and sweat absorption changing with temperature and relative humidity of body or environment. Moreover, nanocomposite application enhanced rigidity, tear strength, anti-felting and weight loss performances of the wool fabric. 20 wt% CNW concentration can be suggested for thermal comfort, mechanical and end use performance enhancements to obtain smart garments having dynamic responsiveness to both body physiological and environmental changes.