In this study, wool fabrics were treated with shape memory polyurethane (SMPU) at different concentrations (5–20 wt%) by using pad-dry-cure process. Transition temperature (Tg) of the SMPU was suitable for body temperature so as to create a fabric having smart breathability and insulation. FT-IR and SEM analyses were conducted for assessing polymer and wool fabric interactions. Air, water vapor permeability (WVP), and water absorption capacity tests of raw and SMPU treated fabrics were carried out within temperature range covering points lower and above Tg of the polymer (20–65 °C). Effects of relative humidity (RH) on WVP were also investigated with the tests carried out under differing RH values (20–80%). According to the results, wool fabrics treated with 10 wt% SMPU had the best smart transfer capability changing according to temperature and relative humidity. Its absorption capacity was also superior with its acceptable hand values according to bending rigidity results.