The effects of chain extender content (ethylene diamine, EDA) and NCO/OH ratio on the properties of natural rubber-based waterborne polyurethanes (WPUs) were investigated experimentally. The particle size of WPU increased significantly with the NCO/OH ratio, in the presence of the EDA chain extender, while it was unaffected by the EDA content. The water uptake of WPU film increased with the EDA content, while the swelling in various solvents decreased. In a thermal analysis, the second decomposition stage of a WPU film increased with the EDA content and with the NCO/OH ratio that also positively affected the dynamic mechanical and mechanical properties. These factors in WPU films had no the effect on the $T_g$. The stress–strain curves clearly showed the change in WPU films from soft elastomeric materials to ductile and hard plastics.