Strong enhancements of outer Van Allen belt electrons have been shown to have a clear dependence on solar wind speed and on the duration of southward interplanetary magnetic field. However, individual case study analyses also have demonstrated that many geomagnetic storms produce little in the way of outer belt enhancements and, in fact, may produce substantial losses of relativistic electrons. In this study, focused upon a key period in August-September 2014, we use GOES geostationary orbit electron flux data and Van Allen Probes particle and fields data to study the process of radiation belt electron acceleration. One particular interval, 13-22 September, initiated by a short-lived geomagnetic storm and characterized by a long period of primarily northward IMF, showed strong depletion of relativistic electrons (including an unprecedented observation of long-lasting depletion at geostationary orbit) while an immediately preceding, and another immediately subsequent, storm showed strong radiation belt enhancement. We demonstrate with these data that two distinct electron populations resulting from magnetospheric substorm activity are crucial elements in the ultimate acceleration of highly relativistic electrons in the outer belt: the source population (tens of keV) that give rise to VLF wave growth; and the seed population (hundreds of keV) that are, in turn, accelerated through VLF wave interactions to much higher energies. ULF waves may also play a role by either reducing inhibiting or enhancing this process through radial diffusion effects. If any components of the inner magnetospheric accelerator happen to be absent, the relativistic radiation belt enhancement fails to materialize. We then generalize this case study to the extended Van Allen Probes mission duration by investigating the direct relationship between southward IMF (and thus enhanced substorm activity) and relativistic electron enhancement events.