Communication systems and radars widely employ sequences of low peak-to-average power ratio (PAR) or unimodulus to meet the hardware constraints and maximize the power efficiency. Numerous works have probed the unimodular sequence design especially attempting to obtain good correlation properties. Regarding channel estimation, however, sequences of such properties do not necessarily qualify for the mission. And tailored unimodular sequences for the specific criterion concerned are more desirable when we have access to the prior knowledge of the channel impulse response. In this paper, we formulate the problem of unimodular sequence design by optimizing minimum mean square error and conditional mutual information, respectively. The problems turn out nonconvex and we develop efficient algorithms based on the majorization-minimization framework with convergence guaranteed. More general, we also examine optimal sequence design with low PAR constraints. Numerical examples demonstrate the improved results of mean square error, signal-to-noise ratio, and conditional mutual information by using our proposed training sequences, with the efficiency of the derived algorithms illustrated.