This thesis is concerned with developing new methodology for the more efficient execution of centroid molecular dynamics simulations — a method based on the theory of imaginary-time path integrals, commonly used in accurate computational prediction of vibrational spectra and other dynamical properties of condensed-phase molecular systems. This is done through the use of machine learning methods to explicitly construct the potential for the centroid. The results obtained through the new methodology are subsequently systematically compared with results obtained through the older, adiabatic, approach to centroid molecular dynamics. This is done for a range of low-dimensional model systems as well as realistic highly-dimensional molecular systems. The differences in these results and the advantages of the new approach are subsequently discussed. The properties of the potential which has been constructed have also been investigated.