The development of cluster molecules is a recent challenge in natural science to control precisely large molecules. We are working on a new benchmark of cluster molecules based on organic chemistry; the central structure of carborane, namely multiborylmethane, shows a unique reactivity in Suzuki–Miyaura cross-coupling reaction. Another strategy for multimetallic complexes exemplifies the efficient construction of chiral catalysts in asymmetric alkylation reaction using organometallic reagents. The representatives in these strategies will be shown in the presentation.