I will give an overview of the challenges that modern electronic structure theory faces in describing strongly correlated chemical systems from molecules to materials. Our latest developments of multireference methods with special focus on using density matrix embedding theory in combination with multireference solvers and its application to understanding the properties and reactivity of electronically excited states and transition metal-containing systems will be discussed.

As an example, in the second part of my talk I will present our latest work on metal-organic frameworks (MOFs). These materials have potential as catalysts for conversion of light alkanes, feedstocks available in large quantities from shale gas that are changing the economics of manufacturing commodity chemicals. Single non-heme Fe(II) ions present as structural moieties in several MOFs (MIL-100, MIL-101, and MIL-808) are identified by Kohn–Sham density functional calculations and multireference calculations as promising catalysts for C–H bond activation, for ethane and methane following the oxidative activation of iron. Through consideration of the full reaction profile leading to the corresponding alcohols, we have identified key changes in the chemical composition of the node that would modulate catalytic activity. The challenge for conventional Kohn–Sham density functional theory in modeling these extended species with open-shell metals will be highlighted, together with possible ways to address them with multireference methods.