The reaction of metal cations with soils varies greatly depending on the mineral suite of the soil and the chemistry of soil clay minerals. In many weathered soils an abundance of Al-hydroxy-interlayered vermiculite (HIV) is present in the clay-size fraction. The Al in the interlayers is potentially quite reactive in terms of sorbing and retaining metal cations such as Cd, Ni and Zn and therefore may play an important role in retaining these compounds. However, little research has been done to investigate the sorption of heavy metals by HIV and the research that has been done has typically used macroscopic approaches. This research addresses the need to investigate such metal-clay interactions while using a molecular and kinetic approach. X-ray absorption fine structure (XAFS) spectroscopy was used to monitor the chemical environment of Ni when reacted with Al-pillared vermiculite over various times. The sorption of Ni increased when vermiculite was Al pillared. The degree of pillar formation, pH, and reaction time all alter the amount of Ni sorbed and the mechanism of Ni sorption.