We have developed Bi-layer molecular conductors based on (alkyl-dihalopyridinium)[Ni(dmit)$_2$]$_2$ (dmit = 1,3-dithiole-2-thione-4,5-dithiolate), in which halogen bonding (halogen···S association) between cations and anions leads to two different kinds of Ni(dmit)$_2$ anion layers in a crystal. It is expected that such a bi-layer system can exhibit novel physical properties, which are not observed in conventional mono-layer molecular conductors. Indeed, (Me-3,5-DIP)[Ni(dmit)$_2$]$_2$ exhibits both metallic conduction and localized spin paramagnetism, resulting from its bi-layer structure, while (Et-2,5-DBrP)[Ni(dmit)$_2$]$_2$ is constructed from two kinds of Mott insulating layers. It was found that (Et-2I-5BrP)[Ni(dmit)$_2$]$_2$ is isostructural with (Et-2,5-DBrP)[Ni(dmit)$_2$]$_2$, however, they are quite different in magnetic behavior at low temperature.

We now focus on alkyl-monohalothiazolium cation to develop new bi-layer Ni(dmit)$_2$ anion radical salts. Since the monohalothiazolium cation is less bulky than the dihalopyridinium cation, denser packing of Ni(dmit)$_2$ anion radicals leading to better conducting property is expected.

We will present crystal and electronic structures and physical properties of a new salt (Et-4BrT)[Ni(dmit)$_2$]$_2$ and related bi-layer Ni(dmit)$_2$ anion radical salts.