Using a scanning tunnelling microscope break-junction technique, we produce 4,4'-bipyridine (44BP) single-molecule junctions with Ni and Au contacts. Electrochemical control is used to prevent Ni oxidation and to modulate the conductance of the devices via nonredox gating - the first time this has been shown using non-Au contacts. Remarkably the conductance and gain of the resulting Ni-44BP-Ni electrochemical transistors is significantly higher than analogous Au-based devices. Ab-initio calculations reveal that this behavior arises because charge transport is mediated by spin-polarized Ni d-electrons, which hybridize strongly with molecular orbitals to form a "spinterface". Our results highlight the important role of the contact material for single-molecule devices and show that it can be varied to provide control of charge and spin transport.