Kitaev-like bond-directional exchange interactions are novel and quite different from the ubiquitous Heisenberg interactions found in most magnets. These kind of interactions open up new possibilities in the exploration and designing of new quantum magnets which can host novel quantum ground states like spin-liquids. However, Kitaev-like in- teractions have remained only a theoretical construct until recently. Honeycomb iridates A_2IrO_3 (A = Na, Li), offer potential realizations of such novel exchange coupling. In this thesis I have synthesized single crystals of A_2TO_3 (A = Na, Li, K, and T = Ir, Ru) and studied their electrical transport, magnetic, and thermal properties. Our work provides several new results: (i) the first thermodynamic evidence of possible fractionalization of electrons in Na_2IrO_3 because of proximity to the Kitaev spin-liquid state, (ii) evidence through magnetic impurity doping, of fragile magnetic order and importance of nearest- neighbour interactions and spin-orbit coupling in deciding the magnetic ground state in Na_2IrO_3 , (iii) a novel method (reactive ion etching) of surface doping Na_2IrO_3 and possibly other layered oxides has been discovered. The surface conductivity of Na_2IrO_3 crystals could be increased by 11 orders of magnitude by varying etching times. The samples which turned metallic show transport anomalies consistent with charge density wave or structural instabilities, (iv) first crystal growth of Li_2RuO_3 is reported where the Xtals crystallize in the $P2_1/m$ structure and show the expected high temperature magneto-structural transition, and (v) design and crystal growth of a new layered honey- comb lattice iridate K_2IrO_3 with an interlayer separation between Ir honeycomb planes which is more than a factor of 2 larger than in Na_2IrO_3 . Magnetic measurements on crys- tals reveal localized effective spin $S = 1/2$ interacting strongly $\theta = -210$ K but without magnetic order down to 1.8 K. Thus K_2IrO_3 is a new Kitaev spin liquid candidate