The nonlinear coupling between an exothermic chemical reaction and a nanowire or nanotube with large axial heat conduction results in a self-propagating thermal wave guided along the nano-conduit. The thermal conduit accelerates the velocity of the resulting reaction wave by rapidly transporting energy to unreacted fuel regions. The reaction wave induces what we term a concomitant thermopower wave of high power density, resulting in an electrical current in the same direction. At up to 7 W/g, this peak power density is larger than that of many present micro-scale power sources (e.g. fuel cells, batteries) and even about seven times greater than that of commercial Li-ion batteries. Thermopower waves also tend to produce monopolar voltage pulses, although conventional thermoelectric theory clearly predicts bipolar voltage. These waves also generate power in excess of the corresponding thermopower and propagate in materials with both high electrical and thermal conductivity. They therefore hold potential to greatly exceed current figures of merit for standard thermoelectric devices.

In this thesis, I have developed the theoretical framework to describe the thermal and chemical profiles of propagating reaction waves, as well as a theory describing electrical power generation during wave propagation. My analysis of the thermal reaction wave yielded a new analytical solution for one-dimensional reaction and thermal diffusion systems with n-th-order kinetics that obviates the need for many perturbation or numerical approaches that have been developed over the past 80 years. I show that a generalized logistic function describes the temperature and concentration profiles within the solid and provides a solution for the velocity of the reaction wave under a wide range of conditions. This approach offers new insight into such problems spanning several fields in science and engineering, including propulsion and self-propagating high-temperature synthesis (SHS) of materials, as well as the dynamics of thermopower waves.

Experimental temperature and voltage measurements of thermopower waves on carbon nanotubes (CNTs) show that they can generate voltage and power in excess of the predictions of the Seebeck effect for the temperature gradient, as much as four times greater power. We hypothesize that the excess thermopower stems from a chemical potential gradient across the CNTs. The fuel (e.g. picramide) and ambient oxygen adsorb and dope the CNTs ahead of the wave, and desorb and react in the hot zone behind the wave front. Using Raman spectroscopy we show that picramide dopes CNTs. Furthermore, the excess thermopower depends on the mass of fuel added (relative to CNT mass), and the chemical potential difference matches the magnitude of the excess thermopower. This model relies on CNT property data already reported in the literature and requires no additional fit parameters. Thus, a major conclusion of this thesis is that coupling to a chemical reaction can boost the performance of thermoelectric materials through the oft-neglected chemical potential term of the drift-diffusion equation defining charge carrier motion.

Another interesting property of thermopower waves is well defined oscillating wave velocity, for certain values of the chemical reaction kinetics and thermal parameters. Voltage measurements in a cyclotrimethylene-trinitramine (fuel)/multiwalled CNT (conduit) system show oscillations with frequencies in the range of 400 to 5000 Hz. These frequencies agree with velocity oscillations predicted by numerical solution of my thermochemical model of the reaction wave, extended to include thermal transport within the conduits. Thermopower waves
could thus find applications as new types of alternating current (AC) batteries and self-powered signal generators, which could easily be miniaturized.

Microelectromechanical systems (MEMS) and wireless sensor networks would benefit from high-power-density thermopower wave generators to enable functions such as communications and acceleration that are currently hampered by large power packs. Additionally, the “self-discharge” rate of thermopower wave generators is extremely low in contrast to electrochemical storage, since their energy is stored in chemical bonds. Thermopower waves thus provide a new class of scalable, stable energy storage devices, and offer the possibility of exceeding the power limitations of conventional thermoelectric devices.