Carbon fiber composites are a critical technology to enable lightweight materials and meet transportation efficiency goals of the future. Despite the maturity of the manufacturing technology, carbon fibers have largely been restricted to high value-added markets. Oak Ridge National Laboratory along with institutional and industrial collaborators have efforts underway to provide a route forward to drastically reduce the cost of carbon fibers while maintaining DOE performance targets. This talk will cover how experimental work on a wide variety of coal and petroleum feedstocks are helping to bolster the connections between molecular structure and final carbon fiber properties. The end goal of this work will be to produce predictive logical frameworks where properties including load to failure, failure mode, stiffness, and resulting microstructure can be directly tied to atomic composition without the need for rigorous testing and development of each feedstock ad nauseum. Specifically, isotropic and mesophase pitch fibers have been produced materials from both coal and petroleum feedstocks have been shown to produce fibers with up to 2.4 GPa strength with 180 GPa modulus following mild carbonization.