289ak Reaction of Young Chars with Oxygen

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Desorption of surface oxides and regeneration of reactive sites has historically been considered the rate controlling steps of char oxidation. Recent studies have suggested that young lignite chars effectively reduce nitrogen oxide levels in fuel rich environments, but their reactivities decrease rapidly and substantially after the volatiles are driven out of the lignite particles. Oxides on these young lignite chars seem to play a dominant role in this process and in a number of other low nitrogen oxide production processes during combustion. Fundamental understandings of the number distributions and strengths of the surface oxides on young chars are important to the development of advanced nitrogen oxide control and combustion technologies. Furthermore, these understandings can lead to advanced computer codes for the simulation and control of various combustion processes. Nevertheless, research on the correlations of reactivity with the characteristics of chars has traditionally been centered on the old chars, or the chars pyrolyzed with a long residence time, typically from one to three hours. Because of these long residence times, it appears that there is an urgent need to enhance our knowledge of chars in the flame region, mainly the chars produced from pyrolysis and combustion with a residence time of a few seconds.

A U-shaped, semi-flow ceramic reactor equipped with a rapid coal-injection port has been designed, fabricated, and operated to accomplish the project goal. Desorption of surface oxides has been carried out at the same site where the char has been produced to minimize the contact of chars with foreign species. Desorption products are analyzed by an online gas chromatograph / mass spectrometer. North Dakota lignite and Illinois No.6 coal were used to produce young char and old char in the reactor at 629 and 1400°C. In the production of young and old chars, oxygen was introduced into the reactor 1 min and 2 hours after 1 gram of coal was injected into the preheated furnace, respectively. The oxidation was conducted with 20% oxygen at 800 ml / min flow rate for 15 second. After oxidation, the gas stream is switched to helium and the reactor is cooled naturally. Desorption products of weakly bound surface oxides during cooling reflects the transient kinetics. Temperature-programmed desorption (TPD) at 6°C / min is then conducted up to 1700°C.

The results suggest that young chars contain more abundant surface oxides than those of old chars over a wide range of TPD temperature. Lignite chars produce more desorption products than their bituminous coal counterparts. More importantly, large amounts of surface oxides are released from all chars between 1100 and 1700°C, an observation that has not been reported before due to the limitation of furnace temperature in the past. This discovery suggests that the existence of a large amount of stable surface oxides form on the basal plan of the carbon structure. Thus, the activation energy of the rate controlling step of char combustion is likely to be much higher than the commonly accepted value, 300 kJ / mol.