Coal Nitrogen Transformation & The Release of Nitrogen Species During Pyrolysis and Combustion in a Bench-Scale Bubbling Fluidised Bed Reactor

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Abstract

The release of nitrogenous species through devolatilisation and combustion of South African coals has been extensively studied in a 50 mm diameter laboratory-scale bubbling fluidised bed reactor. The tests were carried out to determine the devolatilisation and combustion behaviour of the coals at temperatures ranging from 740 to 980°C. The fluidised bed was operated continuously with coal being fed directly into a bed of sand fluidized by nitrogen. Coal contains about 1-2% nitrogen by weight, of which most the nitrogen in the coal matrix exists as organically bound N-containing heteroatomic ring structures. Various conventional and advanced analyses techniques were performed on the parent coals, produced chars and tars for composition and structure. Nitrogen functional forms in coal, char and tar exists as pyrrolic, pyridinic, quaternary and oxidised &/or protonated nitrogen. X-ray photoelectron spectroscopy (XPS) was used to analyse the transformation of nitrogen functional forms from the parent coal to the respective chars and tars. Pyrrolic nitrogen was the predominant functional form in the parent coal and tars produced at 740°C; pyridinic, quaternary and protonated/oxidised nitrogen were observed to increase in the chars and tars as the temperature increased. Quaternary nitrogen was the prevalent form at the higher end temperatures. HCN and NH₃ are intermediately formed during combustion and have been identified as the two main precursors of nitrogen oxides in several studies. The emission of nitrogen oxides from combustion processes is of major environmental concern. Almost all combustion processes lead to the formation of oxides of nitrogen, emitted as nitric oxide (NO), nitrous oxide (N₂O) and nitrogen dioxide (NO₂). The yields of HCN, NH₃, NO, N₂O, NO₂ and tar released during the devolatilisation and combustion were measured and are reported for the specified temperature range. Increasing temperature resulted in a steady decline of ammonia concentration. The same trend was observed for HCN release but to a very small extent. Nitric oxide and nitrogen dioxide were released in very minute concentrations during devolatilisation. The measured oxides of nitrogen from Lethabo coal displayed distinct decreasing trend with increasing temperature, while products from Glisa coal displayed an erratic trend.

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