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气体再燃低NO_X燃烧中NO与NH_i的反应机理研究

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Research on reaction mechanism between NO and NH_i during low NO_x gas reburning

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摘要 采用量子化学密度泛函理论(DFT)对NO与NH_i自由基的反应机理进行了研究,并结合经典过渡态理论对各反应速率常数进行了计算。结果表明,NO与NH_2自由基的反应体系可通过六个反应通道形成N_2+H_2O、N_2O+H_2和N_2H+OH。从能量变化和反应速率两方面考虑,产物N_2+H_2O最容易生成,其最佳反应通道为$NO+NH_2 \rightarrow\rightarrow N_2+H_2O$;NO与NH自由基的反应体系可通过七个反应通道形成N_2+OH、N_2O+H和N_2H+O;其中,N_2+OH最容易生成,最佳反应通道为$NO+NH \rightarrow\rightarrow N_2+OH$。比较发现, NH比$NH_2$自由基更易与NO发生反应生成$N_2$。因此,在实际运行中改变操作条件,实现$NH_2$等向NH方向转化,有利于$NO_x$的还原。

关键词: NO　NH_i自由基　密度泛函理论(DFT)　反应速率

Abstract: The reaction mechanisms between free radicals NO and NH_i were studied using quantum chemical density functional theory (DFT) and rate constant for each reaction was calculated combining with classical transition state theory in this paper. The results show that there are six different reaction channels for free radicals NO and NH_2, and the products are N_2+H_2O, N_2O+H_2 and N_2H+OH. The optimum reaction channel is $NO+NH_2 \rightarrow\rightarrow N_2+H_2O$ from the views of energy change and reaction rate. Similarly, N_2+OH, N_2O+H and N_2H+O will be produced by seven different reaction channels between free radicals NO and NH, and the optimum reaction channel is $NO+NH \rightarrow\rightarrow N_2+OH$. So it is easier for free radical NH reacting with NO to generate N_2 than NH_2. Therefore, an important conclusion was got that the lower NO_x emission could be obtained by making NH_2 turn to NH in the actual operation.

Key words: NO　free radical NH_i　density functional theory (DFT)　reaction rate

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