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论文

煤及显微组分在超高温开放体系实验中动力学参数确立与天然气形成过程预测

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摘要：

通过对塔里木盆地煤及其显微组分以1 K/min升温速率在开放体系下进行热模拟实验(Tmax=1 200 ℃),确定了CH4与N2生成动力学参数。 根据化学反应动力学原理,当煤岩生成CH4和N2为25个一级平行反应且具有相同频率因子(Ao=1.0×1013 s-1)时,生成CH4与N2每个反应对应的活化能可以通过实验数据与理论计算拟合,获得Gaussian型活化能分布特征为：CH4在活化能为42~90 kcal/mol内,煤、镜质组、壳质组、丝质组和半丝质组生成CH4的主频活化能分别为60、52、50、70和66 kcal/mol；N2在活化能为60~108 kcal/mol内,其主频活化能分别为74、108、100、108和102 kcal/mol。根据这些实验动力学参数,推测了煤岩在地质升温速率为5.3 K/Ma时CH4的转化率：CH4在地质温度为20 ℃时开始形成;当地质温度为160 ℃时,煤岩中CH4已生成总量的80%。镜质组和壳质组CH4转化率要高于煤岩。

关键词： 塔里木盆地; 煤岩与显微组分; 热模拟实验; 活化能; 化学动力学

Determination of kinetic parameters in open system non isothermal pyrolysis with ultra high temperature for coal and its macerals and geological extrapolation of natural gas

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Abstract:

Abstract: Open system non isothermal pyrolysis experiments at a heating rate of 1 K/min were performed on immature coal samples (Ro=0.4%) from the Tarim Basin, NW China, and on their isolated macerals (vitrinite, exinite, fusinite and semi fusinite) in order to determine the kinetics of gas generation (CH4 and N2). Assuming that the generation rate of CH4 and N2 was described by 25 single first order reactions with a single pre exponential (Ao＝1.0×1013 s-1), a discrete distribution of activation energies was computed by data processing for CH4 and N2. When the activation energies were distributed between 42 and 90 kcal/mol, a model occurred with the dominent activation energy of 60, 52, 50, 70, and 66 kcal/mol for CH4 from coal, vitrinite, exinite, fusinite and semi fusinite, respectively; the dominant activation energy for N2 was 74, 108, 100, 108 and 102 kcal/mol respectively when the activation energies were in the range of 60108 kcal/mol. Based on the kinetic parameters derived from laboratory experiments, CH4 transformation ratios were calculated for geological heating rate of 5.3 K/Ma. Gas containing more than 80% of CH4 was formed under these conditions at temperatures in excess of 160 ℃ from coal, whereas the percentage of methane generation from vitrinite and exinite was higher than that from coal.

Keywords:

Key words: Tarim Basin; coal and maceral; utra high temperature pyrolysis; activation energy; kinetic modelling

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