Développement d'un outil de détermination de cinétiques par microcalorimétrie différentielle en flux continu : Application aux réactions catalytiques hétérogènes

Doctorant·e

MATEO ROSADO YAMILY

Direction de thèse

POLAERT ISABELLE (Directeur·trice de thèse)
BALLAND LAURENT (Co-directeur·trice de thèse)
LEDOUX ALAIN (Co-directeur·trice de thèse)

Date de la soutenance

13/12/2024 à 09:00

Lieu de la soutenance

LSPC-INSA ROUEN - Avenue de l'Université 76800 Saint Etienne du Rouvray

Rapporteurs de la thèse

FONGARLAND PASCAL Université Claude Bernard - Lyon 1
POSTOLE GEORGETA IRCELYON, Villeurbanne

Membres du jurys

BALLAND LAURENT, , Université de Rouen Normandie (URN)
BEURROIES ISABELLE, , Université d'Aix-Marseille, Faculté des Sciences.
FONGARLAND PASCAL, , Université Claude Bernard - Lyon 1
LEDOUX ALAIN, , INSA de Rouen Normandie
POLAERT ISABELLE, , INSA de Rouen Normandie
POSTOLE GEORGETA, , IRCELYON, Villeurbanne

Abstract

This thesis proposes a new experimental methodology based on differential scanning calorimetry (DSC) in a continuous flow configuration, for the determination of kinetic parameters in chemical reactions. The methodology provides a rapid and accurate estimate of these parameters by measuring the heat released or absorbed during the reaction. The use of a continuous-flow DSC calorimeter enables us to work with small quantities of sample and to carry out the reaction in the measurement zone, offering the advantage of accurately measuring the heat power generated, while controlling temperature and pressure conditions. The methodology details all the steps to be followed and outlines the critical points to be examined and validated if the kinetic study is to be valid. This methodology was applied to the catalytic hydrogenation of CO2 to methane, a rapid exothermic process, using a Ni/Al2O3 catalyst. This reaction was used to assess the importance of the choice of reactor model for obtaining the kinetic parameters by considering the measurement zone as a Continuous Stirred Tank Reactor (CSTR) or a plug flow reactor. The kinetic parameters, such as pre-exponential factors, activation energies and reaction orders, were estimated using a genetic algorithm that minimizes the difference between the experimental thermal power and the calculated thermal power. The methodology therefore makes it possible to select from several kinetic models the one that is most representative of the experimental results. The methodology also includes an analysis of the sensitivity of the parameters to the accuracy of the kinetic model. The results obtained show good agreement between the experimental data and the model predictions, validating the effectiveness of this methodology for the kinetic analysis of heterogeneous reactions in continuous flow. This approach represents an effective alternative to traditional methods, offering new possibilities in the optimization of catalytic processes and the study of complex systems. This thesis presents an innovative tool based on differential calorimetry in continuous flow, capable of generating reliable kinetic data, facilitating progress in the field of catalysis and chemical kinetics