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On the Effects of Reactant Flow Rarefaction on Heterogeneous Catalysis: a Regularized Lattice Boltzmann Study

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Abstract. In this paper, a numerical investigation on heterogeneous catalysis is performed by means of a Regularized Lattice BGK approach. The effects of different values of the reactant flow Knudsen numbers are evaluated, in terms of conversion efficiency and penetration inside the structure of a nano-porous gold ingot. The results are in line with experimental evidence in the literature and open interesting perspectives for the optimal design of future nano-catalytic devices.

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1 Introduction

The investigation of the activity of solid catalysts is at the heart of chemical engineering and physical chemistry for the broad range of applications and technological processes related to such phenomenon, from material synthesis to pollutant treatment, from fuel production to conversion processes in general [6].

This is the reason why theoretical and computational studies of the interactions at gas-solid interfaces are central to further optimization of complex real-life catalytic processes. At a fundamental level, detail models are available for *high-vacuum* experiments, those characterized by a pressure of $\sim 10^{-3} \div 10^{-4}$ *torr*, corresponding to Knudsen (Kn) values

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for the gas flow much larger than the unity [8,14]. In [7], transport in the Knudsen regime of inert gases through straight channels was considered: it was shown how small-scale surface geometry of a macroscopically flat channel affects the diffusion process of the gas within the porous structure of the catalyst.

The Lattice Boltzmann Method derives from an optimized version of Boltzmann's Kinetic Equation, thus consisting of successive free flight processes along characteristics and collisional relaxation towards a Maxwellian equilibrium of a set of discrete distribution functions [1,3–5,9,10,15]. Due to its kinetic foundations, the Lattice Boltzmann Method (LBM) has proven to provide an efficient tool for the investigation of heterogeneous catalysis at $Kn \lesssim 1$ [2,6,12]. It has been also recently shown [11] that the use of the LB equation augmented with regularization procedure, namely a Hermite projection of post-collision distribution functions, allows for the simulation of fluid flows across a wide range of Knudsen numbers. Exploiting the capability of LB to deal with such complex fluid phenomena, in this paper, we delve into the characteristics of gas-solid interactions in nano-porous gold catalysts exposed to reactant fluxes at different Kn numbers. The results show a significant dependence of catalytic reaction distribution on Kn parameter, as expected from experimental works in the literature, confirming the reliability of LBM for flow regimes characterized by moderate vacuum levels.

2 Numerical methodology

In this work, we employ a multicomponent lattice Boltzmann model augmented with suitable reactive boundary conditions [6,12]. The multicomponent lattice Boltzmann equation reads as follows:

$$f_i^{(k)}(\boldsymbol{x}+\boldsymbol{c}_i,t+1)-f_i^{(k)}(\boldsymbol{x},t)=-\omega(f_i^{(k)}-f_i^{(k)eq}), \tag{2.1}$$

where $f_i^{(k)}$ is the probability density function of finding a particle of the species k at site $\boldsymbol{x}$ at time t, moving along the $i-th$ lattice direction defined by the discrete speeds $\boldsymbol{c}_i$, with $i=0,\cdots,b$, where $b=8$ if a two dimensional nine speed lattice (D2Q9) is employed.

The left hand-side of Eq. (2.1) represents the free-streaming of molecules, whereas the right-hand side accounts for the collisional relaxation towards local Maxwellian equilibrium, on a time-scale $\tau=1/\omega$. The macroscopic fluid density ρ and velocity $\boldsymbol{u}$ are given by $\rho(\boldsymbol{x},t)=\sum_{i=0}^b f_i(\boldsymbol{x},t)$ and $\rho(\boldsymbol{x},t)\boldsymbol{u}(\boldsymbol{x},t)=\sum_{i=0}^b \boldsymbol{c}_i f_i(\boldsymbol{x},t)$, respectively. The equilibrium distribution function is given by a low-Mach, second-order, expansion of a local Maxwellian, namely:

$$f_i^{k,eq}=w_i\rho^k\left(1+\frac{1}{c_s^2}\boldsymbol{c}_i\cdot\boldsymbol{u}+\frac{1}{2c_s^4}(\boldsymbol{c}_i\cdot\boldsymbol{u})^2-\frac{1}{2c_s^2}\boldsymbol{u}\cdot\boldsymbol{u}\right), \tag{2.2}$$

where c_s is the lattice speed of sound and w_i is the standard set of weight of the D2Q9 stencil [15]. In order to investigate the effect of rarefaction on catalytic processes inside