Comparison of gas excess adsorption models and high pressure experimental validation

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A B S T R A C T

The adsorption data of the pure gases methane, ethylene and hydrogen on two types of zeolites and two types of activated carbon materials were measured using a volumetric method at 293 K and at pressure up to 26.8 MPa. Four different approaches were tested for description of the experimental equilibrium data. Based on the experimental data obtained by other authors at very wide range of pressure (0–650 MPa), we propose a new modeling type adsorption isotherm, describing the excess of adsorption of supercritical gases on micro porous adsorbents. The analysis was developed for the description of the phenomena minimum of adsorption isotherms at very high pressure. The concept of inflection point and its thermodynamically treatments were developed. The comparison of the different models describing adsorption isotherms allows a critical discussion of their accuracy.

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1. Introduction

A gas physisorption phenomenon is very important aspect especially in gas mixture separation industry (Barto et al., 1984). Data for adsorption equilibrium at very high pressure range are required in the design of gas separation and purification equipment. They are important also from a scientific point of view. However, there are few studies of adsorption on microporous adsorbents at very high pressure. At these conditions, over 10 MPa and ambient temperature, most of the gases are in supercritical state. Thus, excess adsorption isotherms differ from typical isotherms according to classification of IUPAC (1985, 1994) itemized by Aranovich and Donohue (1998) and Donohue and Aranovich (1998). The authors proposed a new classification for excess adsorption isotherms at these specific conditions. It is considered that there is a maximum in the excess adsorption isotherm (Aranovich and Donohue, 1996a,b, 1998; Do, 1998; Do and Do, 2003; Donohue and Aranovich, 1998a; Hocker et al., 1999; Yang, 1988) with the increase of pressure. Data presented by both Malbrunot et al. (1992) and Menon (1965, 1968), show that the isotherms of adsorption exhibit an adsorption minimum at very high pressure.

Based on the experimental data (Malbrunot et al., 1992; Menon, 1965, 1968) and on the theoretical results of Darkrim et al. (1999), and Chilev et al. (2009) we propose a new type of adsorption isotherm and give the simple analysis treatment of the minimum of adsorption isotherms that appear at very high pressure.

Few theories and models have been developed to correlate adsorption data at relatively high pressures. Among them, the model proposed by Do and Do (Chilev et al., 2009; Darkrim et al., 1999; Do, 1998; Do and Do, 2003, 2005; Hocker et al., 1999; Malbrunot et al., 1992; Menon, 1965, 1968; Yang, 1988), a two-dimensional equation of state (Zhou et al., 1994), Monte Carlo simulations (Do and Do, 2005; Weinberger, 2001; Zhou et al., 1994), the classical lattice theory applying in the case of microporous adsorbents (Aranovich et al., 2000; Aranovich and Donohue, 1996a,b, 1998; Donohue and Aranovich, 1998b; Hocker et al., 1999; Ono and Kondo, 1960), the three parameter isotherm equation (Beutekamp, 2002; Dreisbach et al., 2002) and the equation of Chilev et al. (2009).

Some of these models present certain advantages than others due to their simplicity and, in some cases, due to their application. Accuracy of an isotherm model is generally due