Phase Behavior of N$_2$O and CO$_2$ in Room-Temperature Ionic Liquids [bmim][Tf$_2$N], [bmim][BF$_4$], [bmim][N(CN)$_2$], [bmim][Ac], [eam][NO$_3$], and [bmim][SCN]

Mark B. Shiflett · Anne Marie S. Niehaus · Beth A. Elliott · A. Yokozeki

Received: 14 June 2011 / Accepted: 27 December 2011 / Published online: 14 February 2012 © Springer Science+Business Media, LLC 2012

Abstract The gas solubility of nitrous oxide (N$_2$O) in room-temperature ionic liquids, 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, 1-butyl-3-methylimidazolium tetrafluoroborate, 1-butyl-3-methylimidazolium dicyanamide, 1-butyl-3-methylimidazolium acetate, 1-butyl-3-methylimidazolium thiocyanate, and ethylammonium nitrate has been measured at isothermal conditions from about (283 to 348) K using a gravimetric microbalance. The observed pressure–temperature composition ($P$,$T$,$x$) data have been analyzed by use of a generic Redlich-Kwong equation-of-state (EOS) model, which has been successfully applied in our previous works. The interaction parameters have been determined using our measured vapor–liquid equilibrium data. Vapor–liquid–liquid equilibrium measurements have been made and validate EOS model predictions which suggest that these systems demonstrate Type III and Type V phase behavior, according to the classification of van Konynenburg and Scott. The global phase behavior of N$_2$O has also been compared with both the measured data from this study and literature data for carbon dioxide (CO$_2$) in each ionic liquid and Henry’s law constants are compared at room temperature (298.15 K).

Keywords Carbon dioxide · Equation of state · Gas separation · Gas solubility · Ionic liquid · Liquid–liquid separation · Nitrous oxide · Phase equilibria

Electronic supplementary material The online version of this article (doi:10.1007/s10765-011-1150-4) contains supplementary material, which is available to authorized users.

M. B. Shiflett (✉) · A. M. S. Niehaus · B. A. Elliott
DuPont Central Research and Development, Experimental Station, Wilmington, DE 19880, USA
e-mail: mark.b.shiflett@usa.dupont.com

A. Yokozeki
32 Kingsford Lane, Spencerport, NY 14559, USA
1 Introduction

The current focus on global warming gases is centered on carbon dioxide emissions; however, nitrous oxide (N$_2$O) is also an important greenhouse gas that is produced from both natural and industrial sources. N$_2$O has an atmospheric lifetime of about 114 years and a global warming potential (GWP) of 298 (100-year time horizon) which, when compared with CO$_2$ (GWP = 1), makes N$_2$O a potent greenhouse gas [1–6]. The atmosphere contains about 319 ppbv of N$_2$O (2005), which is a 9% increase from pre-industrial levels (285 ppbv) with an annual growth rate of about 0.2% to 0.3% [2–4]. N$_2$O is one of six greenhouse gases (CO$_2$, CH$_4$, N$_2$O, HFC, PFC, SF$_6$) identified for reduction by the United Nations Framework on Climate Change (UNFCC) [4].

The primary source of N$_2$O is from biological processes in soils and oceans which represent about two-thirds of the total emissions [2,4,7]. Anthropogenic sources of N$_2$O include agricultural activities and industries such as mobile and stationary combustion, nitric and adipic acid production, and waste-water treatment [4,7]. A number of industries have voluntarily initiated efforts to reduce N$_2$O emissions, particularly from adipic acid production. Thermal and catalytic abatement technologies have been successfully developed for this application due to the high N$_2$O concentration in the adipic acid tail-gas (typically 25 vol% to 40 vol%) [4]. However, these technologies are not applicable to other sources such as nitric acid plants or stationary combustion processes due to dissimilar characteristics of the tail gas (i.e., lower N$_2$O concentrations). If N$_2$O can be efficiently separated from adipic acid tail-gas (20 vol% N$_2$O, 20 vol% NO$_2$, plus CO$_2$, O$_2$, and N$_2$) it could be recycled to produce additional adipic acid. Therefore, there still exists the need for development of an efficient and economic technology for the removal of N$_2$O.

A possible solution is to selectively recover N$_2$O on a sorbent and produce a concentrated stream of N$_2$O during desorption. Attempts to concentrate these streams by selective adsorption using metal-exchanged zeolites has led to N$_2$O concentrations of 5 vol% which is still insufficient to satisfy practical requirements [4,8]. Recently, room-temperature ionic liquids (RTILs) have been proposed for the capture and separation of N$_2$O. Anthony et al. [9] reported the first solubility data of N$_2$O in 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([bmim][Tf$_2$N]) at three temperatures (283.15, 298.15, 323.15) K and pressures up to 1.3 MPa. They show that the solubility of N$_2$O and CO$_2$ are essentially the same (both mole and mass fraction basis) in [bmim][Tf$_2$N] and have a significantly higher solubility than other hydrocarbons (C$_2$H$_4$, C$_2$H$_6$) and oxygen. Revelli et al. [10] measured the solubility of N$_2$O in five different imidazolium-based ionic liquids with a variable volume view cell at temperatures from (293 to 373) K and pressures up to 30 MPa. They claim that 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF$_4$]) and 1,3-diethloxyimidazolium bis(trifluoromethylsulfonyl)imide (([ETO]$_2$IM)[Tf$_2$N]) are the most efficient ionic liquids for capturing N$_2$O and also show that the type of fluid phase behavior is Type III according to the classification of Scott and van Konynenburg [11,12].