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Synthesis of polymeric ionic liquids material and application in CO_2 adsorption

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We synthesized one quaternary ammonium polymeric ionic liquids (PILs)P[VBTHEA]Cl and three imidazolium PILs of P[VEIm]Br, P[VEIm]BF_4, P[VEIm]PF_6 by free-radical polymerization in solution. These PILs were characterized by FT-IR, ^{1}H-NMR, ^{13}C-NMR, TGA, XRD and SEM. Their CO_2 adsorption capacities were measured under different pressures and temperatures by constant-volume technique. It was observed that quaternary ammonium PILs of P[VBTHEA]Cl have higher adsorption capacity for CO_2 than those imidazolium PILs, following P[VBTHEA]Cl > P[VEIm]PF_6 > P[VEIm]BF_4 > P[VEIm]Br, which may be ascribed to higher positive charge density on ammonium cation than that on imidazolium cation and thus stronger interaction with CO_2, consistent with the results from dual-mode adsorption model that ammonium PILs have much higher CO_2 bulk absorption than imidazolium PILs. CO_2 adsorption capacity of P[VBTHEA]Cl is 9.02 mg/g under 295 K and 1 bar, which is comparable to that of some other PILs, and is much higher than that of the corresponding ILs monomer. These PILs have good adsorption selectivity for CO_2 over N_2 and regeneration efficiency.

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1. Introduction

Excessive consumption of fossil fuels has caused high CO_2 accumulation in atmosphere and aroused greenhouse effect, which results in a series of serious environmental problems such as glacier melting, sea level rising, ocean acidification and extreme weather. CO_2 concentration in atmosphere has increased from 315 ppm in 1958 to 391 ppm in 2011, and close to 398 ppm in 2014 [1]. The pH value of surface sea water is expected to drop from 8.2 to 7.8 by 2095 [2]. It is very urgent to reduce CO_2 emission. Carbon capture and storage (CCS) is an effective technology for mitigating this problem. CO_2 capture technologies include absorption, adsorption, membrane separation, cryogenic distillation, etc. [3–5], among which alkanolamine solution absorption technology is a mature process. High energy consumption and corrosion are typically problems in the alkanolamine solution process [6,7].

Room-temperature ionic liquids, entirely composed of organic cations and organic/inorganic anions, are liquid organic salts at or close to room temperature, also called ionic liquids (ILs) in short. Compared with traditional solvents, ILs have a lot of unique advantages such as extremely low vapor pressure, good chemical and thermal stability, wide electrochemical potential window [8]. Many researchers have observed that CO_2 is highly soluble in imidazolium ILs. CO_2 solubility in [BMIM]PF_6 was 0.75 (molar fraction) at 8.3 MPa [9]. CO_2 solubility in a series of imidazolium ILs with $[BMIM]^+$ and different anions followed $[NO_3]^- < [N(CN)_2]^- < [BF_4]^- < [PF_6]^- < [CF_3SO_3]^- < [NTf_2]^-$ [10]. Higher CO_2 solubility was observed in those ILs with fluo-

Abbreviations: BIEMA, 2-(1-butylimidazolium-3-yl)ethyl methacrylate; BIEO, (1-butylimidazolium-3)methylethyleneoxide; BMIM, 1-butyl-3-methylimidazolium; EEIM, 1,3-bis(2-methoxy-2-oxyl ethyl) imidazolium; HHIM, 1,3-bis(2-hydroxyl ethyl) imidazolium; MABI, 1-[2-(methacryloyloxy)ethyl]-3-butyl-imidazolium; MBA, N,*N*-methylenebisacrylamide; Met, methioninate; NTf_2, bis(trifluoromethylsulfonyl)imide; OTf, trifluoromethanesulfonate; P66614, trihexyl(tetradecyl)phosphonium; Pro, prolinate; Sac, *o*-benzoicsulphimide; VBBI, 1-(Ivinylbenzyl)-3-butyl-imidazolium; VBMI, 1-(Ivinylbenzyl)-3-methyl-imidazolium; VBTBA, *p*-vinylbenzyltributylammonium; VBTEA, *p*-vinylbenzyltriethylammonium; VBTHEA, 4-vinylbenzyl triethanolamine; VBTMA, *p*-vinylbenzyltrimethylammonium; VEIm, *N*-vinylimidazole.

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Table 1. CO_2 adsorption capacity of various types of PILs.

PILs	CO_2 loading (mg/g)	Conditions pressure (bar)/temperature (K)	Ref.
P[VBTBA]BF_4	3.5	0.78/295	[18]
P[VBTMA]NTf_2	2.74	0.78/295	[18]
P[VBTMA]Sac	3.27	0.78/295	[18]
P[VBTEA]BF_4	6.99	0.78/295	[18]
P[VBTEA]PF_6	10.36	1298	[31]
MBA-crosslinked- P[VBTEA]PF_6	14.04	1298	[31]
P[VBMI]BF_4	4.64	0.78/295	[24]
P[VBBI]BF_4	3.20	0.78/295	[24]
P[VBBI]PF_6	3.16	0.78/295	[24]
P[VBBI]NTf_2	1.87	0.78/295	[24]
P[VBBI]Sac	1.59	0.78/295	[24]
P[MABI]BF_4	2.49	0.78/295	[24]
P[BIEO]BF_4	1.72	0.78/295	[24]
P[HHIm-EEIM]BF_4	4.54	0.86/298	[33]
P[HHIm-EEIM]PF_6	3.35	0.86/298	[33]
P[BIEMA]BF_4 (MW, 19,000)	2.41	0.78/295	[23]
P[BIEMA]BF_4 (MW, 6708)	2.99	1298	[34]
P[BIEMA]PF_6 (MW, 7862)	3.31	1298	[34]
P[BIEMA]NTf_2 (MW, 10,543)	1.53	1298	[34]
P[BIEMA]OTf (MW, 7943)	2.05	1298	[34]
P[BIEMA]Br (MW, 6571)	2.88	1/298	[34]
P[VBTHEA]Cl	9.02	1295	This work
P[VEIm]Br	4.15	1295	This work
P[VEIm]BF_4	5.26	1295	This work
P[VEIm]PF_6	5.63	1295	This work

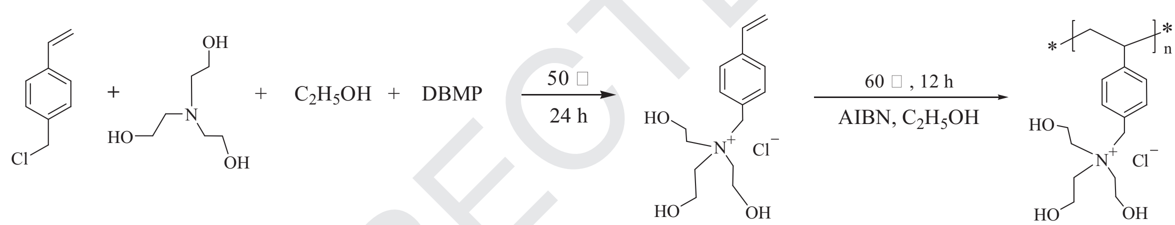

Fig. 1. Synthetic route of P[VBTHEA]Cl.

rine element in anion, which was called fluorination [11]. Solubility of CO_2 in [BMIM]PF_6 is much higher than that of C_2H_4, C_2H_6, CH_4, Ar, O_2, CO, H_2 and N_2 [12].The solubility of CO_2 in these common ILs is physical and not high in lower pressure, e.g., atmospheric pressure. Some task-specific ILs, with importing functional groups, were prepared to enhance CO_2 absorption capacity, e.g., [P_{66614}]Met, [P_{66614}]Pro where CO_2 absorption capacity was close to 1 mol_{CO_2}/mol_{IL} [13]; unfortunately, high viscosity is usually observed after absorbing CO_2. Recently, some solid adsorbents such as polymeric ionic liquids (PILs), carbon nanotubes [14] and MOFs [15] have been tested, among which PILs have been attracting more and more attention.

PILs are obtained through the polymerization of ILs monomer with polymerizable groups such as vinyl and styryl. The polymers of ILs monomer, PILs, are solid polymer materials in most circumstances, and not liquid while we call them PILs. PILs have similar properties to ILs monomer for their same functional group, while the former, with polymer structure, is designable and machinable. PILs are suitable in some applications of gas adsorption separation, electrochemistry, catalyst, biotechnology, etc. [16,17]. It has been observed that PILs have very good CO_2 adsorption and separation capacity. Tang et al. [18–24] synthesized several PILs of imidazolium, quaternary ammonium, quaternary phosphonium and pyridinium; CO_2 adsorption capacities of these PILs were much better than that of the corresponding ILs monomers, and adsorption/desorption was fast and reversible; CO_2 adsorption capacity followed quaternary ammonium > pyridinium > quaternary phosphonium > imidazolium. The CO_2 adsorption of ammonium-based PILs is generally higher than that of imidazolium- and pyridinium-based PILs, owing to the presence of sp^3 hybridization in the ammonium cations that can readily rearrange and become more accessible to CO_2 thus facilitating CO_2 sorption in contrast to other ILs having sp^2 hybridization in the aromatic cation groups [25]; on the other hand, there is higher microvoid volume fraction in ammonium-based PILs than that in imidazolium-based PILs [21]. In addition, Tang et al. [21] observed that the anions also play an important role in the sorption of CO_2, e.g., the CO_2 sorption capability follows this order $P[VBTMA][BF_4] > P[VBTMA][PF_6] >> P[VBTMA][Tf_2N]$ and the much lower CO_2 sorption capability of P[VBTMA]Tf_2N may be due to the lower microvoid fraction and weaker interaction with CO_2 from the plasticization by the anion. Supasitmongkol and Styring [26] synthesized quaternary ammonium PILs of P[VBTMA]PF_6 and observed that it can adsorb 77% of its body weight of CO_2 with selectivity over N_2 of 70:1. Shahrom et al. [27] prepared AAPILs with functionalized amine on anion and the AAPILs absorbed 0.53 fraction of CO_2 at 0.7 bar and 298 K. Bara et al. [28] prepared ILs–PILs separation membranes with 400% increase of CO_2 permeability and 33% improvement of CO_2/N_2 selectivity. Qiu et al. [29] prepared Pebax1657 membranes with [BMIM]NTf_2, and CO_2 permeability reached up to approximately 286 Barrer with the addition of [BMIM]NTf_2. Porous polymeric microparticles of P[VBTEA]Cl, P[VBTEA]BF_4 and P[VBTEA]PF_6 were prepared by us via inverse suspension polymerization, and good CO_2 adsorption capacity was observed [30,31]. Two review papers are available for CO_2 adsorption by PILs [25,32], and some results are listed in Table 1 [18,23,24,31,33,34].

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