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Semiclathrate Hydrate Phase Equilibrium for CO_2/CH_4 Gas Mixtures in the Presence of Tetrabutylammonium Halide (Bromide, Chloride, or Fluoride)

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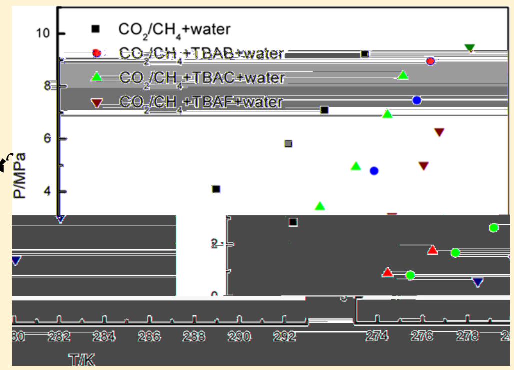
K L E H E C , M E , C E , C , G 510640, C

ABSTRACT: I CO_2/CH_4 + w , CO_2/CH_4 + (BAB) + w , CO_2/CH_4 + (BAC) + w , CO_2/CH_4 + fl (BAF) + w 280.2 K 291.3 K 0.61 M 9.45 M w 2.93·10^{-3} w 0.33 CO_2 0.67 CH_4. CH_4 + w . B w BAB, BAC, BAF. A , BAF BAF > BAC > BAB. CO_2/CH_4 + BAF + w 0.61 M 284.2 K.



INTRODUCTION

G w ( ) w ( ) G (CH_4, CO_2, C_2H_6, N_2, H_2) w w I (I), II (II), H(H), w [1]

B w w F w [2] (:), (BAB), (BAC), fl (BAF). I (BA^+) w (B $^-$, Cl$^-$, F$^-$) w w H- [3] [4–8] O [9–12] F BAB w [13–15] M [16–21] fl) w ff CO_2, CH_4, N_2, H_2,

N [22–24] w A w w w CH_4 CO_2. O CO_2 CH_4 CO_2 [25] (273 ffi K 283 K 1 M 6 M) w CH_4 CO_2 w A BA(B, C, F) ff ffi D D [26] CH_4/CO_2 w 0.4 BAB. w BA BA H w BA CO_2 [27]

I w CO_2/CH_4 (0.33 0.67) w w (2.93·10^{-3} (, fl). w

EXPERIMENTAL SECTION

Materials. 1

R : J 23, 2013
A : 19, 2013
P : O 8, 2013

1. E M

CH_4	0.9999 ( )	G	C .
CO_2/CH_4	0.33 0.67 ( )	G	C .
BAB	0.99 ( )	K	C .
BAC	0.99 ( )	K	C .
BAF	0.97 ( )		C .

2.93·10^{-3} (w_{BAB} = 0.050, w_{BAC} = 0.043, w_{BAF} = 0.049). 0.33 CO_2 0.67 CH_4 ± 0.003.

BAB, BAC, BAF 0.01.

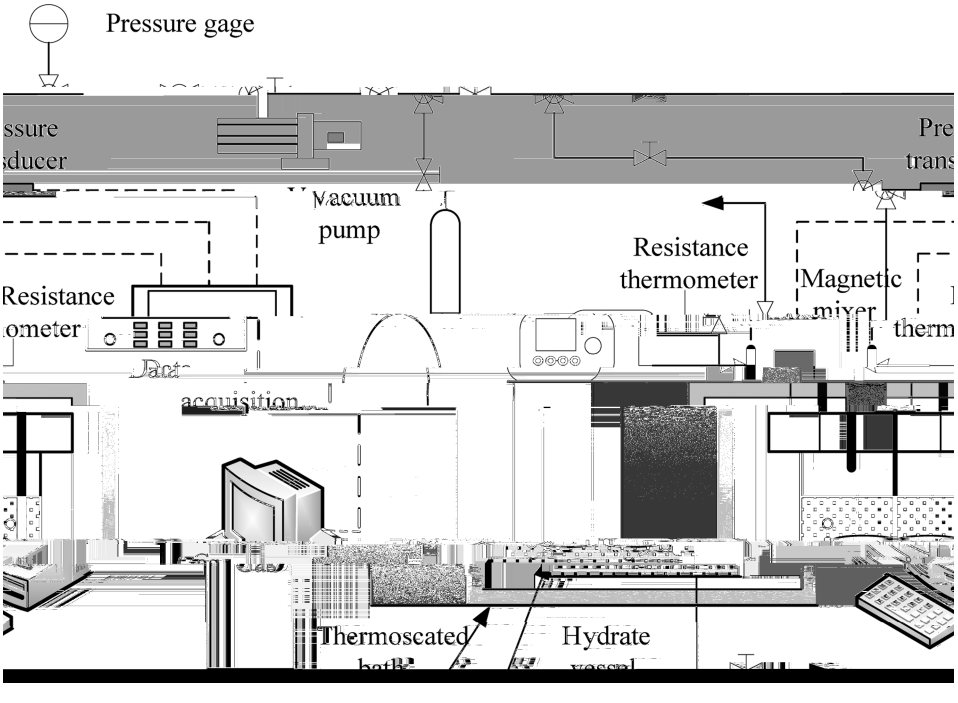
Experimental Apparatus. F 1 [16] 60 80 ff 300 ^{3}. D , fl (GBM 76 024) I , 20 M , (H CC2-K20B) (- 100) ± 0.1 K . (DG-1300) ± 0.01 M . All (A 34970A) 20 ,

CO_2/CH_4 (GC).

Experimental Method. [28,29] F , 200 L , CO_2/CH_4 , GC, . A 30 ( ), , 3 K/ H . A 0.1 K. fl 4 . , *P*–*T* . I ,

RESULTS AND DISCUSSION

CH_4 + H_2O [30] 2 3. F , F 2. I F 2 ,



F 1.

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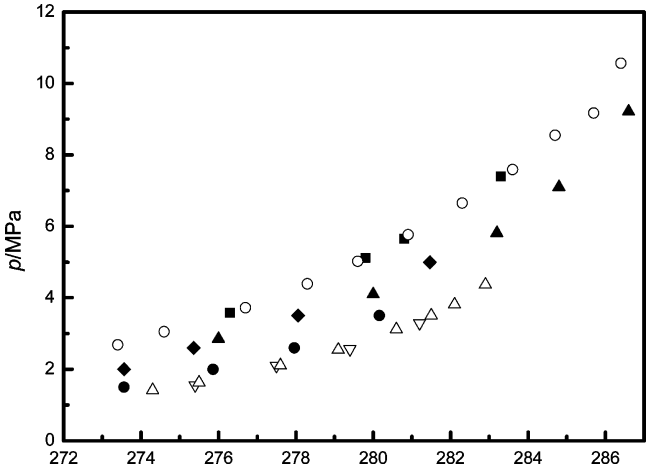
2. M H F C CH_4

T/K	p/M
276.3	3.58
279.8	5.12
281.4	6.05
283.3	7.39

3. M H F C CO_2/CH_4

T/K	p/M
276.0	2.85
280.0	4.10
283.2	5.82
284.8	7.10
286.6	9.22



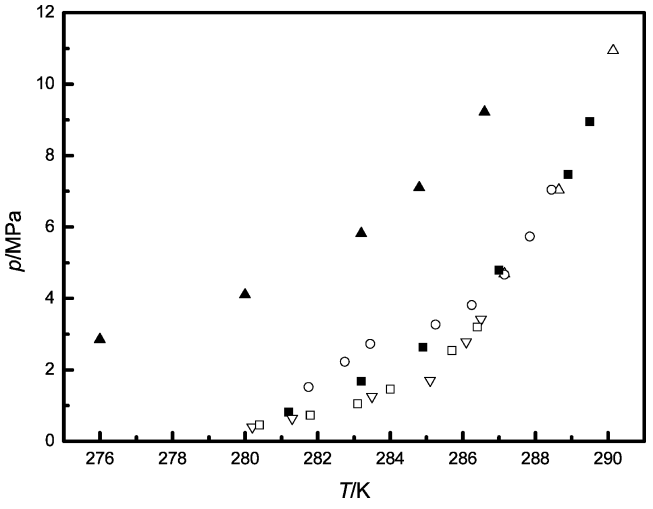
F 2. $CH_4 + H_2O$, $CO_2/CH_4 + H_2O$ $CO_2 + H_2O$. $CH_4 + H_2O$: ■, ; ○, 30. $CO_2/CH_4 + H_2O$: ♦, 1 (CO_2, $y = 0.20$); ▲, (CO_2, $y = 0.33$); ●, 1(CO_2, $y = 0.60$). $CO_2 + H_2O$: ▽, 16; △, 0.

I , CH_4/CO_2 (3) $CH_4 +$ $CO_2 +$ 0 6. I F 2, CO_2/CH_4 $CH_4 +$ $CO_2 +$. $CO_2/CH_4 +$ BAB + (4) CH_4

4. H E D CO_2+CH_4+ BAB + S M 2.93·10^{-3} M F BAB

T/K	p/M
282.0	0.78
284.6	1.65
286.2	2.60
288.3	4.76
290.2	7.44
291.1	8.94

+ BAB + $CO_2 +$ BAB + $CO_2/CH_4 +$ (F 3).



F 3. $CH_4 +$ BAB + , $CO_2 +$ BAB + , $CO_2/CH_4 +$ $CO_2/CH_4 +$ BAB + . $CH_4 +$ BAB + : △, 2; ○, 3. $CO_2 +$ BAB + : ▽, 6; □, 9. $CO_2/CH_4 +$: ▲, . $CO_2/CH_4 +$ BAB + : ■, .

BAB II 2.93·10^{-3} . F 3, $CO_2/CH_4 +$ BAB + $CH_4 +$ BAB + $CO_2 +$ BAB + 4 M ; 4 M , $CO_2/CH_4 +$ BAB + $CH_4 +$ BAB + . I BAB (2.93·10^{-3}).

, 5 6 $CO_2 + CH_4 +$ BAC + $CO_2 + CH_4 +$ BAF

5. H E D $CO_2 + CH_4 +$ BAC + S M 2.93·10^{-3} M F BAC

T/K	p/M
280.2	0.90
282.2	1.75
284.6	3.42
286.2	4.94
287.6	6.92
288.3	8.40

+ . F 4 5 $CO_2 + CH_4 +$ BAC + $CO_2 + CH_4 +$ BAF + $CO_2 + CH_4 +$. BAC BAF (2.93·10^{-3}). A F 3 F 5 $CO_2 +$ $CH_4 +$ BAF + , CO_2

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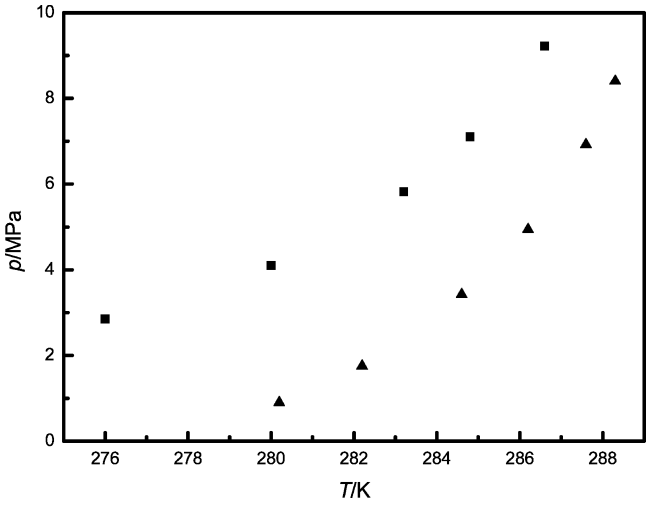
6. H E D $CO_2 + CH_4 +$ BAF + S M $2.93 \cdot 10^{-3}$ M F BAF

T/K	p/M
284.2	0.61
285.8	1.45
287.8	3.07
289.2	5.02
289.9	6.30
291.3	9.45



F 4. CO

+ CH_4 + BAF + 5−11 K $CO_2 + CH_4 +$.

CONCLUSION

CO_2 (0.33) + CH_4 (0.67) + : $CO_2 + CH_4 +$ BAB + , $CO_2 + CH_4 +$ BAC + $CO_2 + CH_4 +$ BAF + ,

280.2 291.3 K 0.61 9.45 M , $x = 2.93 \cdot 10^{-3}$. . A $CO_2 + CH_4 +$ + $CO_2 + CH_4 +$ (BAB, BAC, BAF) . A 285 K, $CO_2 + CH_4 +$ (BAB, BAC, BAF) + 3.74 M , 2.76 M 0.94 M , , 7.30 M $CO_2 + CH_4 +$. A , BAF , BAF > BAC > BAB.

AUTHOR INFORMATION

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Notes

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