Supplementary Information

Chinese herbs: treasure troves for the discovery of environmentally friendly promoters for methane hydrate formation

Suying Wang, Yanxian Zeng, Yuanhao Cai, Xiaochun Niu, Zhengmei Zhu, Dongjun Lei and Weixing Wang*

Key Laboratory of Enhanced Heat Transfer and Energy Conservation of the Ministry of Education, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou, Guangdong 510640, China.

* Correspondence to: cewxwang@scut.edu.cn

Materials

Chinese herbs were purchased from Guangdong Jinkang Pharmacy Chain Co., Ltd., China. Magnesium hydroxide (99%), baicalin (95%), amygdalin (97%), methyl gallate (98%), and sodium dodecyl sulfate (SDS, 97%) were purchased from Aladdin Co. Ltd., China. Methane (99.9%) was purchased from Zhuozheng Gas Co. Ltd., China.

High-throughput experiments

2.0 g of each dried Chinese herb (353 K overnight) was infused in 100 g of water at a temperature of 353 K for 60 min (eight samples as one group). The extracted suspension was filtered to obtain a clear solution (Chinese herbal extract solution). The residue was dried at 353 K over night to calculate the extraction rate and concentration of the Chinese herbal extract solution. 1.0 g of each Chinese herbal extract solution was placed in a separate glass vial (4 mL). To carry out the methane uptake kinetic high-throughput experiments, eight sample solutions as one group in a holder were loaded into a high-pressure stainless-steel vessel with a volume of 120 cm^3 (Kerui Instruments Co., Gongyi, Henan, China). The temperature of the coolant was controlled by using a programmable thermal circulator (DWHW-15, Kerui Instruments Co., Gongyi, Henan, China). Prior to each experiment, the cell was purged with methane three times to remove the air, and then pressurized to the starting pressure of 9.5 MPa (273 K). After reacting for 5 h under isochoric condition at 273 K, the excess methane was rapidly vented. The amount of methane uptake was measured via volumetric displacement of water at room temperature (298 ± 3 K) and 1 atm.

Synthesis of magnesium baicalin

0.2 g of baicalin and 0.013 g of magnesium hydroxide were reacted in water under stirring at 60 ℃ for 2 h to obtain a clear solution of magnesium baicalin.

Methane Hydrates Formation in a Static Reactor[1]

To perform the methane uptake kinetic experiments, sample solution (20.0 g) was loaded into a high-pressure stainless-steel vessel with a volume of 82 cm^3 (Kerui Instruments Co., Gongyi, Henan, China). The temperature of the coolant was controlled by using a programmable thermal circulator (DWHW-10, Kerui Instruments Co., Gongyi, Henan, China). The sample temperature in the high-pressure cell was measured using a type K thermocouple (−250 to 400 °C, Tianyi Cekong Co., Henan, China). The methane gas pressure was monitored using a high-accuracy gauge pressure transmitter (0–20 MPa, Tianyi Cekong Co., Henan, China). Both the thermocouple and the transmitter were connected to a digital universal input panel meter (ZNHW, Kerui Instruments Co., Gongyi, Henan, China), to communicate with the computer. Prior to each experiment, the cell was purged with methane three times to remove the air, and then pressurized to the desired pressure (starting pressure was 9.5 MPa unless otherwise specified) at the designated temperature. The temperature (T), pressure (P), and time (t) were automatically interval-logged using SuperCx 3.0 software (Kerui Instruments Co., Gongyi, Henan, China).

Calculation of methane uptake kinetics[1, 2]

The methane storage capacity was defined gravimetrically in terms of the mass of absorbed of methane per unit mass of the aqueous solution at 273 K. Capacities were calculated using the pressure change that occurs in the reaction vessel. The free space volume of the vessel was obtained by subtracting the sum volume of methane clathrate hydrate, unreacted water, and the additive. Taking into account non-ideality factors, GASPAK v3.41 software (Horizon Technologies, USA) was employed to calculate the gas enclathration capacity. We assume that the liquid and gas phases inside the vessel were exclusively formed from the solution and the guest gas, respectively, neglecting any dissolution of the guest gas into the liquid phase and any mixing of the water vapor in the gas phase.

Table S1. Extraction rates, concentration of extracts and storage capacities of 300 kinds of Chinese herbs.

No.	Latin name*	Chinese name*	Extraction rate (%)†	Concentration of extract (wt%)‡	Methane uptake (mL g^{-1})§
1	*Abri Herba*	Jigucao	5.6	0.11	0
2	*Acanthopanacis Senticosi Radix Et Rhizoma Seu Caulis*	Ciwujia	5.0	0.10	0
3	*Achyranthis Bidentatae Radix*	Niuxi	62.5	1.23	80
4	*Aconiti Kusnezoffii Radix*	Caowu	19.0	0.38	0
5	*Aconiti Kusnezoffii Radix Cocta*	Zhicaowu	27.5	0.55	0
6	*Aconiti Lateralis Radix Praeparata*	Fuzi	14.4	0.29	0
7	*Aconiti Radix*	Chuanwu	13.4	0.27	0
8	*Aconiti Radix Cocta*	Zhichuanwu	19.0	0.38	0
9	*Adenophorae Radix*	Nanshashen	29.9	0.59	2
10	*Agrimoniae Herba*	Xianhecao	13.9	0.28	204
11	*Akebiae Caulis*	Mutong	7.1	0.14	0
12	*Akebiae Fructus*	Yuzhizi	39.6	0.79	0
13	*Albiziae Flos*	Hehuanhua	13.4	0.27	0
14	*Allii Macrostemonis Bulbus*	Xiebai	35.5	0.70	34
15	*Allii Tuberosi Semen*	Jiucaizi	6.0	0.12	0
16	*Aloe*	Luhui	65.6	1.30	0
17	*Alpiniae Officinarum Rhizoma*	Gaoliangjiang	8.5	0.17	0
18	*Ampelopsis Radix*	Bailian	19.0	0.38	0
19	*Andrographis Herba*	Chuanxinlian	15.4	0.31	0
20	*Anemones Raddeanae Rhizoma*	Liangtoujian	17.6	0.35	0
21	*Angelicae Dahuricae Radix*	Baizhi	13.7	0.27	194
22	*Angelicae Pubescentis Radix*	Duhuo	55.3	1.09	0
23	*Angelicae Sinensis Radix*	Danggui	69.8	1.38	18
24	*Apocyni Veneti Folium*	Luobumaye	25.8	0.51	5
25	*Arctii Fructus*	Niubangzi	3.4	0.07	0
26	*Ardisiae Japonicae Herba*	Aidicha	8.0	0.16	0
27	*Arecae Pericarpium*	Dafupi	13.6	0.27	50

28	*Arecae Semen*	Binglang	5.9	0.12	0
29	*Arisaematis Rhizoma Preparatum*	Zhitiannanxing	8.5	0.17	0
30	*Aristolochiae Fructus*	Madouling	15.0	0.30	56
31	*Armeniacae Semen Amarum*	Kuxingren	28.5	0.57	0
32	*Artemisiae Annuae Herba*	Qinghao	16.0	0.32	0
33	*Artemisiae Argyi Folium*	Aiye	16.5	0.33	0
34	*Asari Radix Et Rhizoma*	Xixin	12.5	0.25	0
35	*Asparagi Radix*	Tiandong	12.3	0.25	0
36	*Asteris Radix Et Rhizoma*	Ziwan	38.0	0.75	20
37	*Astragali Complanati Semen*	Shayuanzi	13.8	0.28	0
38	*Astragali Radix*	Huangqi	23.5	0.47	0
39	*Atractylodis Macrocephalae Rhizoma*	Baizhu	54.6	1.08	22
40	*Atractylodis Rhizoma*	Cangzhu	31.7	0.63	0
41	*Aucklandiae Radix*	Muxiang	38.2	0.76	205
42	*Aurantii Fructus*	Zhiqiao	38.5	0.76	0
43	*Aurantii Fructus Immaturus*	Zhishi	17.5	0.35	0
44	*Bambusae Caulis In Taenias*	Zhuru	6.9	0.14	0
45	*Bambusae Concretio Silicea*	Tianzhuhuang	9.6	0.19	0
46	*Belamcandae Rhizoma*	Shegan	13.0	0.26	0
47	*Benincasae Exocarpium*	Dongguapi	23.6	0.47	0
48	*Benzoinum*	Anxixiang	63.2	1.25	0
49	*Bletillae Rhizoma*	Baiji	60.2	1.19	210
50	*Broussonetiae Fructus*	Chushizi	6.1	0.12	0
51	*Bruceae Fructus*	Yadanzi	15.9	0.32	0
52	*Buddlejae Flos*	Mimenghua	22.8	0.45	0
53	*Bupleuri Radix*	Chaihu	14.0	0.28	0
54	*Campsis Flos*	Lingxiaohua	29.1	0.58	0
55	*Carotae Fructus*	Nanheshi	17.7	0.35	0
56	*Carthami Flos*	Honghua	37.3	0.74	180
57	*Caryophylli Flos*	Dingxiang	14.6	0.29	0
58	*Celosiae Cristatae Flos*	Jiguanhua	24.5	0.49	174

59	*Celosiae Semen*	Qingxiangzi	5.0	0.10	0
60	*Centellae Herba*	Jixuecao	25.2	0.50	158
61	*Centipedae Herba*	Ebushicao	38.2	0.76	0
62	*Chaenomelis Fructus*	Mugua	21.6	0.43	0
63	*Changii Radix*	Mingdangshen	7.3	0.15	5
64	*Chebulae Fructus*	Hezi	23.8	0.47	210
65	*Chelidonii Herba*	Baiqucai	32.0	0.64	0
66	*Chuanxiong Rhizoma*	Chuanxiong	46.7	0.93	0
67	*Cibotii Rhizoma*	Gouji	32.9	0.65	40
68	*Cimicifugae Rhizoma*	Shengma	59.0	1.17	30
69	*Cinnamomi Ramulus*	Guizhi	12.3	0.25	0
70	*Cirsii Japonici Herba*	Daji	43.4	0.86	170
71	*Cistanches Herba*	Roucongrong	29.2	0.58	0
72	*Citri Fructus*	Xiangyuan	33.2	0.66	0
73	*Citri Grandis Exocarpium*	Huajuhong	39.4	0.78	0
74	*Citri Reticulatae Pericarpium*	Chenpi	8.9	0.18	0
75	*Citri Reticulatae Pericarpium Viride*	Qingpi	18.4	0.37	0
76	*Citri Reticulatae Semen*	Juhe	8.9	0.18	0
77	*Citri Sarcodactylis Fructus*	Foshou	50.0	0.99	0
78	*Clematidis Armandii Caulis*	Chuanmutong	12.3	0.25	0
79	*Clematidis Radix Et Rhizoma*	Weilingxian	14.8	0.30	0
80	*Cnidii Fructus*	Shechuangzi	44.3	0.88	0
81	*Codonopsis Radix*	Dangshen	25.7	0.51	0
82	*Commelinae Herba*	Yazhicao	11.9	0.24	0
83	*Coptidis Rhizoma*	Huanglian	20.4	0.41	20
84	*Corni Fructus*	Shanzhuyu	37.2	0.74	0
85	*Corydalis Decumbentis Rhizoma*	Xiatianwu	12.4	0.25	15
86	*Crataegi Fructus*	Shanzha	23.5	0.47	0
87	*Cremastrae Pseudobulbus, Pleiones Pseudobulbus*	Shancigu	21.5	0.43	0
88	*Curculiginis Rhizoma*	Xianmao	14.0	0.28	0
89	*Cuscutae Semen*	Tusizi	18.5	0.37	0

90	*Cyathulae Radix*	Chuanniuxi	27.2	0.54	0
91	*Cynanchi Atrati Radix Et Rhizoma*	Baiwei	6.1	0.12	0
92	*Cynanchi Paniculati Radix Et Rhizoma*	Xuchangqing	21.8	0.43	0
93	*Cynanchi Stauntonii Rhizoma Et Radix*	Baiqian	11.2	0.22	0
94	*Cynomorii Herba*	Suoyang	18.1	0.36	0
95	*Cyperi Rhizoma*	Xiangfu	6.3	0.13	0
96	*Dendrobii Caulis*	Shihu	8.6	0.17	0
97	*Descurainiae Semen, Lepidii Semen*	Tinglizi	7.6	0.15	0
98	*Dianthi Herba*	Qumai	7.0	0.14	0
99	*Dictamni Cortex*	Baixianpi	9.7	0.19	0
100	*Dioscoreae Nipponicae Rhizoma*	Chuanshanlong	16.8	0.33	0
101	*Dipsaci Radix*	Xuduan	33.3	0.66	0
102	*Drynariae Rhizoma*	Gusuibu	7.6	0.15	0
103	*Ecliptae Herba*	Mohanlian	15.1	0.30	198
104	*Ephedrae Herba*	Mahuang	16.7	0.33	0
105	*Epimedii Folium*	Yinyanghuo	9.3	0.19	0
106	*Equiseti Hiemalis Herba*	Muzei	14.9	0.30	0
107	*Eriobotryae Folium*	Pipaye	6.0	0.12	0
108	*Erodii Herba Geranii Herba*	Laoguancao	13.9	0.28	0
109	*Erycibes Caulis*	Dinggongteng	6.0	0.12	0
110	*Eucommiae Cortex*	Duzhong	10.0	0.20	0
111	*Euodiae Fructus*	Wuzhuyu	21.3	0.42	0
112	*Eupatorii Herba*	Peilan	16.0	0.32	0
113	*Euphorbiae Hirtae Herba*	Feiyangcao	13.1	0.26	0
114	*Euryales Semen*	Qianshi	10.9	0.22	0
115	*Farfarae Flos*	Kuandonghua	47.0	0.93	215
116	*Ferulae Resina*	Awei	7.7	0.15	0
117	*Fibraureae Caulis*	Huangteng	7.5	0.15	0
118	*Forsythiae Fructus*	Lianqiao	12.5	0.25	0
119	*Fraxini Cortex*	Qinpi	11.3	0.23	0

120	*Galangae Fructus*	Hongdoukou	5.0	0.10	0
121	*Galla Chinensis*	Wubeizi	48.5	0.96	74
122	*Gardeniae Fructus*	Zhizi	13.6	0.27	0
123	*Gastrodiae Rhizoma*	Tianma	36.3	0.72	0
124	*Gendarussae Herba*	Xiaobogu	5.9	0.12	0
125	*Gentianae Macrophyllae Radix*	Qinjiao	9.9	0.20	0
126	*Gentianae Radix Et Rhizoma*	Longdan	19.6	0.39	0
127	*Ginkgo Folium*	Yinxingye	16.0	0.32	0
128	*Ginseng Folium*	Renshenye	38.0	0.75	0
129	*Gleditsiae Fructus Abnormalis*	Zhuyazao	14.1	0.28	0
130	*Gleditsiae Spina*	Zaojiaoci	3.7	0.07	205
131	*Glehniae Radix*	Beishashen	15.2	0.30	0
132	*Gossampini Flos*	Mumianhua	15.3	0.31	5
133	*Granati Pericarpium*	Shiliupi	17.5	0.35	0
134	*Hibisci Mutabilis Folium*	Mufurongye	17.5	0.35	108
135	*Homalomenae Rhizoma*	Qiannianjian	10.9	0.22	0
136	*Houttuyniae Herba*	Yuxingcao	16.6	0.33	0
137	*Ilicis Chinensis Folium*	Sijiqing	16.4	0.33	0
138	*Ilicis Rotundae Cortex*	Jiubiying	13.1	0.26	0
139	*Imperatae Rhizoma*	Baimaogen	13.5	0.27	0
140	*Inulae Flos*	Xuanfuhua	14.5	0.29	0
141	*Isatidis Folium*	Daqingye	13.6	0.27	187
142	*Isatidis Radix*	Banlangen	20.0	0.40	0
143	*Junci Medulla*	Dengxincao	6.4	0.13	0
144	*Kaempferiae Rhizoma*	Shannai	10.7	0.21	0
145	*Kaki Calyx*	Shidi	5.1	0.10	0
146	*Kansui Radix*	Gansui	22.2	0.44	0
147	*Kochiae Fructus*	Difuzi	11.9	0.24	0
148	*Lablab Semen Album*	Baibiandou	5.9	0.12	0
149	*Laminariae Thallus, Eckloniae Thallus*	Kunbu	13.5	0.27	0
150	*Lasiosphaera Calvatia*	Mabo	9.5	0.19	0

151	*Leonuri Herba*	Yimucao	16.9	0.34	0
152	*Ligustici Rhizoma Et Radix*	Gaoben	14.5	0.29	0
153	*Lilii Bulbus*	Baihe	25.0	0.50	0
154	*Litchi Semen*	Lizhihe	2.3	0.05	0
155	*Lobeliae Chinensis Herba*	Banbianlian	29.8	0.59	20
156	*Lonicerae Japonicae Flos*	Jinyinhua	39.8	0.79	0
157	*Lophatheri Herba*	Danzhuye	11.4	0.23	0
158	*Luffae Fructus Retinervus*	Sigualuo	8.3	0.17	0
159	*Lycii Folium*	Gouqiye	27.7	0.55	14
160	*Lycii Cortex*	Digupi	5.6	0.11	0
161	*Lycopi Herba*	Zelan	17.2	0.34	52
162	*Lycopodii Herba*	Shenjincao	11.7	0.23	0
163	*Magnoliae Flos*	Xinyi	7.4	0.15	0
164	*Magnoliae Officinalis Flos*	Houpohua	11.6	0.23	0
165	*Menthae Haplocalycis Herba*	Bohe	31.5	0.63	190
166	*Microctis Folium*	Buzhaye	11.6	0.23	0
167	*Momordicae Semen*	Mubiezi	10.0	0.20	0
168	*Mori Cortex*	Sangbaipi	9.2	0.18	0
169	*Mori Folium*	Sangye	30.7	0.61	0
170	*Mori Fructus*	Sangshen	47.6	0.94	0
171	*Mori Ramulus*	Sangzhi	5.0	0.10	0
172	*Morindae Officinalis Radix*	Bajitian	71.9	1.42	0
173	*Moslae Herba*	Xiangru	13.0	0.26	0
174	*Moutan Cortex*	Mudanpi	16.8	0.33	0
175	*Mume Fructus*	Wumei	60.6	1.20	0
176	*Murrayae Folium Et Cacumen*	Jiulixiang	17.0	0.34	0
177	*Myrrha*	Moyao	18.5	0.37	0
178	*Nardostachyos Radix Et Rhizoma*	Gansong	17.0	0.34	0
179	*Nelumbinis Folium*	Heye	11.6	0.23	0
180	*Nelumbinis Plumula*	Lianzixin	35.8	0.71	10
181	*Nelumbinis Rhizomatis Nodus*	Oujie	26.5	0.53	3
182	*Nelumbinis Stamen*	Lianxu	30.1	0.60	42

183	*Notoginseng Radix Et Rhizoma*	Sanqi	20.6	0.41	0
184	*Notopterygii Rhizoma Et Radix*	Qianghuo	53.7	1.06	0
185	*Olibanum*	Ruxiang	17.0	0.34	30
186	*Ophiopogonis Radix*	Maidong	36.0	0.71	20
187	*Paeoniae Radix Alba*	Baishao	16.2	0.32	0
188	*Paeoniae Radix Rubra*	Chishao	12.7	0.25	0
189	*Paridis Rhizoma*	Chonglou	15.2	0.30	0
190	*Perillae Caulis*	Zisugeng	3.2	0.06	0
191	*Perillae Folium*	Zisuye	22.0	0.44	0
192	*Perillae Fructus*	Zisuzi	4.4	0.09	0
193	*Periplocae Cortex*	Xiangjiapi	21.0	0.42	0
194	*Persicae Semen*	Taoren	3.5	0.07	0
195	*Peucedani Radix*	Qianhu	13.2	0.26	0
196	*Pharbitidis Semen*	Qianniuzi	24.0	0.48	0
197	*Phellodendri Chinensis Cortex*	Huangbo	14.7	0.29	0
198	*Phragmitis Rhizoma*	Lugen	9.0	0.18	0
199	*Physalis Calyx Seu Fructus*	Jindenglong	19.9	0.40	0
200	*Phytolaccae Radix*	Shanglu	32.6	0.65	0
201	*Pini Pollen*	Songhuafen	43.2	0.86	0
202	*Piperis Kadsurae Caulis*	Haifengteng	16.0	0.32	0
203	*Piperis Longi Fructus*	Bibo	6.4	0.13	0
204	*Plantaginis Herba*	Cheqiancao	24.9	0.50	220
205	*Plantaginis Semen*	Cheqianzi	9.9	0.20	5
206	*Platycladi Cacumen*	Cebaiye	12.7	0.25	0
207	*Platycladi Semen*	Baiziren	11.3	0.23	0
208	*Platycodonis Radix*	Jiegeng	60.4	1.19	0
209	*Pogostemonis Herba*	Guanghuoxiang	17.2	0.34	0
210	*Polygalae Radix*	Yuanzhi	23.1	0.46	0
211	*Polygonati Odorati Rhizoma*	Yuzhu	68.7	1.36	0
212	*Polygonati Rhizoma*	Huangjing	49.7	0.98	0
213	*Polygoni Avicularis Herba*	Bianxu	6.6	0.13	0

214	*Polygoni Cuspidati Rhizoma Et Radix*	Huzhang	7.9	0.16	150
215	*Polygoni Multiflori Caulis*	Shouwuteng	15.4	0.31	40
216	*Polygoni Perfoliati Herba*	Gangbangui	8.0	0.16	0
217	*Polyporus*	Zhuling	39.4	0.78	0
218	*Poriae Cutis*	Fulingpi	7.4	0.15	0
219	*Portulacae Herba*	Machixian	16.5	0.33	205
220	*Potentillae Discoloris Herba*	Fanbaicao	8.3	0.17	42
221	*Prunellae Spica*	Xiakucao	9.6	0.19	198
222	*Pruni Semen*	Yuliren	8.7	0.17	0
223	*Pseudolaricis Cortex*	Tujingpi	43.5	0.86	0
224	*Pseudostellariae Radix*	Taizishen	29.4	0.58	72
225	*Psoraleae Fructus*	Buguzhi	6.3	0.13	0
226	*Puerariae Lobatae Radix*	Gegen	25.0	0.50	0
227	*Pulsatillae Radix*	Baitouweng	13.1	0.26	156
228	*Pyrolae Herba*	Luxiancao	8.8	0.18	0
229	*Pyrrosiae Folium*	Shiwei	14.0	0.28	0
230	*Quisqualis Fructus*	Shijunzi	14.0	0.28	0
231	*Rabdosiae Rubescentis Herba*	Donglingcao	8.2	0.16	0
232	*Ranunculi Ternati Radix*	Maozhaocao	18.4	0.37	0
233	*Raphani Semen*	Laifuzi	11.1	0.22	0
234	*Rehmanniae Radix*	Dihuang	65.4	1.29	40
235	*Rehmanniae Radix Praeparata*	Shudihuang	42.0	0.83	0
236	*Rhapontici Radix*	Loulu	13.2	0.26	220
237	*Rhei Radix Et Rhizoma*	Dahuang	25.0	0.50	35
238	*Rhodiolae Crenulatae Radix Et Rhizoma*	Hongjingtian	10.9	0.22	186
239	*Rosae Chinensis Flos*	Yuejihua	11.8	0.24	52
240	*Rosae Laevigatae Fructus*	Jinyingzi	45.4	0.90	0
241	*Rosae Rugosae Flos*	Meiguihua	20.7	0.41	210
242	*Rubi Fructus*	Fupenzi	8.7	0.17	0
243	*Rubiae Radix Et Rhizoma*	Qiancao	16.4	0.33	0
244	*Salviae Miltiorrhizae Radix Et Rhizoma*	Danshen	56.6	1.12	190

245	*Sanguisorbae Radix*	Diyu	15.1	0.30	205
246	*Saposhnikoviae Radix*	Fangfeng	17.2	0.34	68
247	*Sappan Lignum*	Sumu	5.6	0.11	0
248	*Sarcandrae Herba*	Zhongjiefeng	12.7	0.25	0
249	*Sargassum*	Haizao	17.7	0.35	0
250	*Sargentodoxae Caulis*	Daxueteng	11.7	0.23	0
251	*Sauropi Folium*	Longliye	32.9	0.65	70
252	*Saururi Herba*	Sanbaicao	10.8	0.22	0
253	*Schisandrae Chinensis Fructus*	Wuweizi	36.0	0.71	0
254	*Schizonepetae Herba*	Jingjie	12.7	0.25	10
255	*Schizonepetae Spica*	Jingjiesui	20.2	0.40	0
256	*Scrophulariae Radix*	Xuanshen	49.1	0.97	20
257	*Scutellariae Barbatae Herba*	Banzhilian	12.8	0.26	142
258	*Scutellariae Radix*	Huangqin	48.6	0.96	200
259	*Sedi Herba*	Chuipencao	16.2	0.32	50
260	*Selaginellae Herba*	Juanbai	11.0	0.22	0
261	*Semiaquilegiae Radix*	Tiankuizi	35.3	0.70	0
262	*Sennae Folium*	Fanxieye	33.2	0.66	182
263	*Siegesbeckiae Herba*	Xixiancao	12.7	0.25	30
264	*Silybi Fructus*	Shuifeiji	5.7	0.11	0
265	*Sinapis Semen*	Jiezi	12.7	0.25	0
266	*Smilacis Glabrae Rhizoma*	Tufuling	8.0	0.16	0
267	*Sojae Semen Germinatum*	Dadouhuangjuan	20.4	0.41	0
268	*Sojae Semen Praeparatum*	Dandouchi	17.0	0.34	0
269	*Solidaginis Herba*	Yizhihuanghua	8.0	0.16	0
270	*Sophorae Flavescentis Radix*	Kushen	12.3	0.25	0
271	*Sophorae Tonkinensis Radix Et Rhizoma*	Shandougen	27.2	0.54	50
272	*Sparganii Rhizoma*	Sanleng	8.7	0.17	0
273	*Spatholobi Caulis*	Jixueteng	5.6	0.11	0
274	*Spirodelae Herba*	Fuping	16.0	0.32	0
275	*Stellariae Radix*	Yinchaihu	24.8	0.49	180

276	*Stemonae Radix*	Baibu	56.6	1.12	0
277	*Stephaniae Tetrandrae Radix*	Fangji	23.0	0.46	0
278	*Sterculiae Lychnophorae Semen*	Pangdahai	21.2	0.42	2
279	*Tamaricis Cacumen*	Xiheliu	5.8	0.12	15
280	*Taraxaci Herba*	Pugongying	33.6	0.67	15
281	*Taxilli Herba*	Sangjisheng	5.8	0.12	0
282	*Tinosporae Radix*	Jinguolan	19.7	0.39	15
283	*Toosendan Fructus*	Chuanlianzi	24.0	0.48	0
284	*Torreyae Semen*	Feizi	2.8	0.06	0
285	*Toxicodendri Resina*	Ganqi	3.1	0.06	0
286	*Trachelospermi Caulis Et Folium*	Luoshiteng	7.4	0.15	0
287	*Tribuli Fructus*	Jili	25.7	0.51	0
288	*Trichosanthis Pericarpium*	Gualoupi	43.0	0.85	0
289	*Trichosanthis Radix*	Tianhuafen	13.7	0.27	0
290	*Trigonellae Semen*	Huluba	14.2	0.28	0
291	*Tsaoko Fructus*	Caoguo	28.3	0.56	0
292	*Typhae Pollen*	Puhuang	17.6	0.35	0
293	*Vaccariae Semen*	Wangbuliuxing	15.8	0.32	0
294	*Valerianae Jatamansi Rhizoma Et Radix*	Zhizhuxiang	18.1	0.36	160
295	*Verbenae Herba*	Mabiancao	16.0	0.32	0
296	*Vignae Semen*	Chixiaodou	5.9	0.12	126
297	*Violae Herba*	Zihuadiding	31.3	0.62	0
298	*Visci Herba*	Hujisheng	21.1	0.42	0
299	*Zanthoxyli Pericarpium*	Huajiao	26.0	0.52	0
300	*Zingiberis Rhizoma*	Ganjiang	11.8	0.24	0

* 《Pharmacopoeia of the People's Republic of China》, 2020 edition.
† The weight percentage of the water-soluble fraction of the dried Chinese herb.
‡ The concentration of the Chinese herbal extract aqueous solution.
§ At room temperature (298 ± 3 K) and 1 atm pressure.

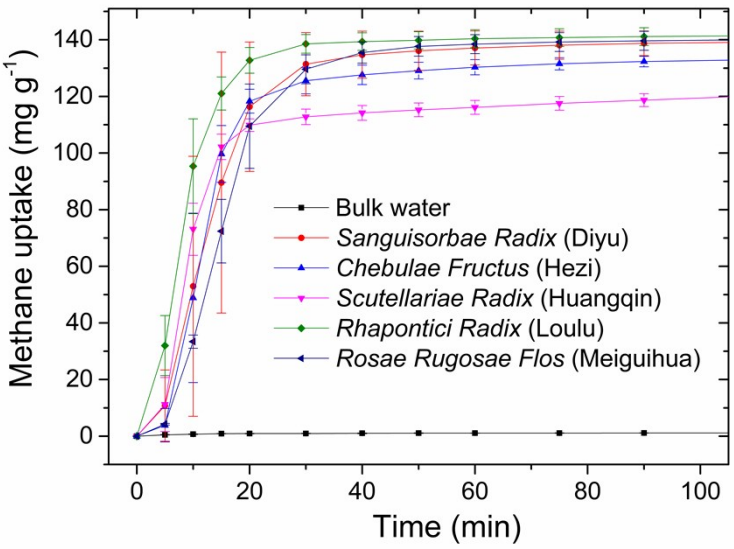

Fig. S1 Methane uptake kinetics for the bulk water and Chinese herbal extract aqueous solutions at 273 K (initial pressure, 9.5 MPa, local enlargement of Fig. 2).

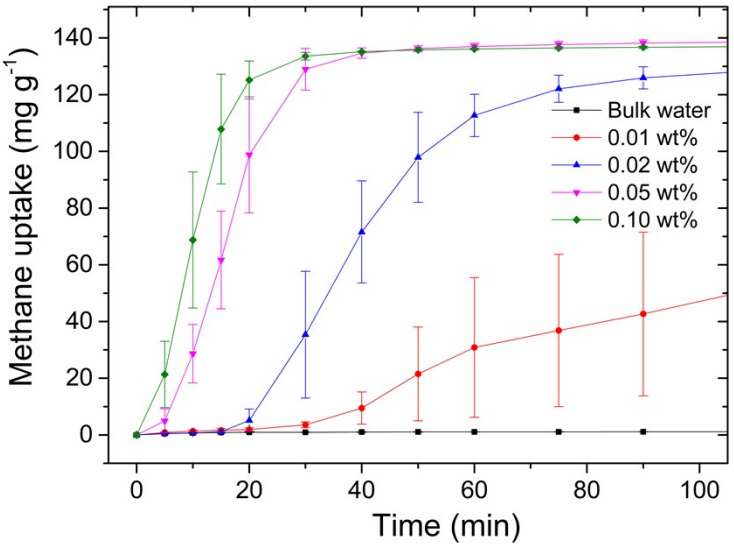

Fig. S2 Methane uptake kinetics for the bulk water and *Scutellariae Radix* extract aqueous solutions at 273 K (initial pressure, 9.5 MPa, local enlargement of Fig. 3).

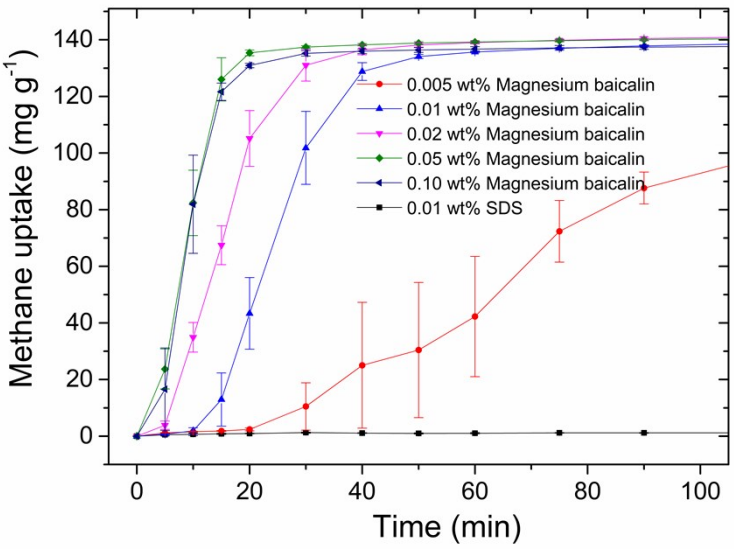

Fig. S3 Methane uptake kinetics for SDS and magnesium baicalin aqueous solutions at 273 K (initial pressure, 9.5 MPa, local enlargement of Fig. 4a).

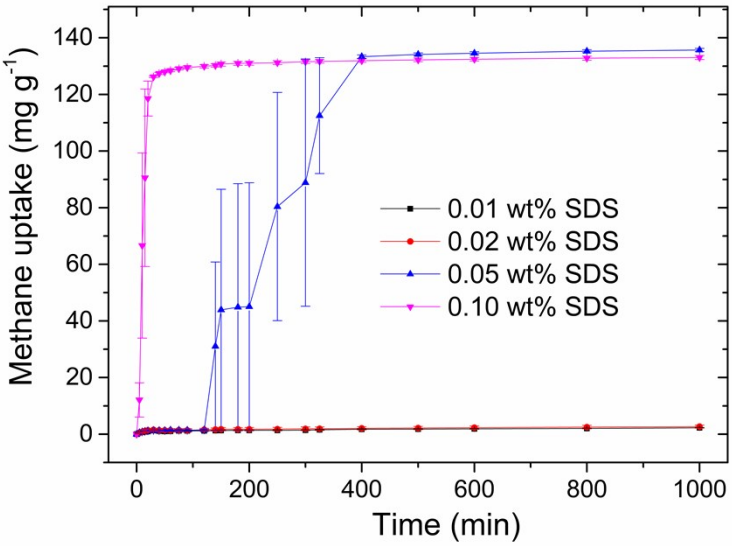

Fig. S4 Methane uptake kinetics for SDS aqueous solutions at 273 K (initial pressure, 9.5 MPa).

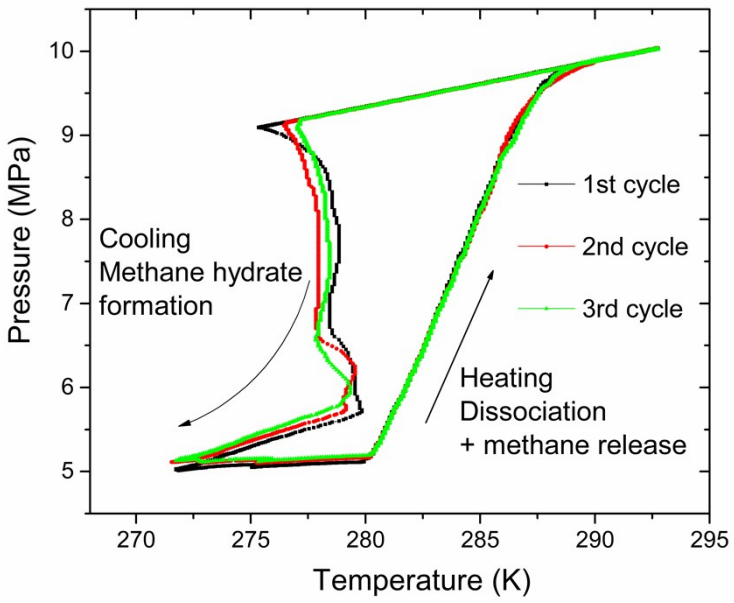

Fig. S5 Recyclability of 0.02 wt% magnesium baicalin aqueous solution during cooling and heating under methane pressure (temperature ramp: 4.0 K h^{-1}).

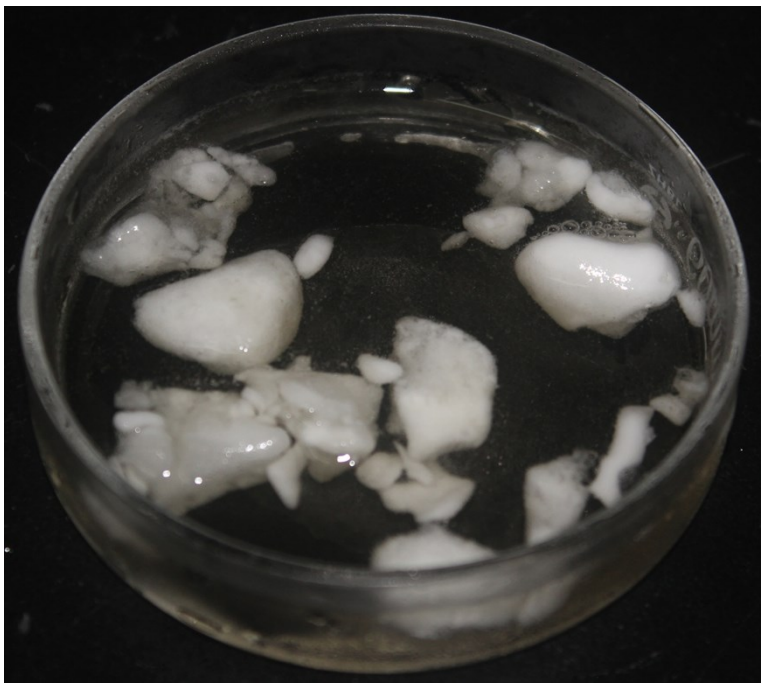

Fig. S6 Methane hydrate (0.02 wt% magnesium baicalin) dissociated without foaming upon degassing.

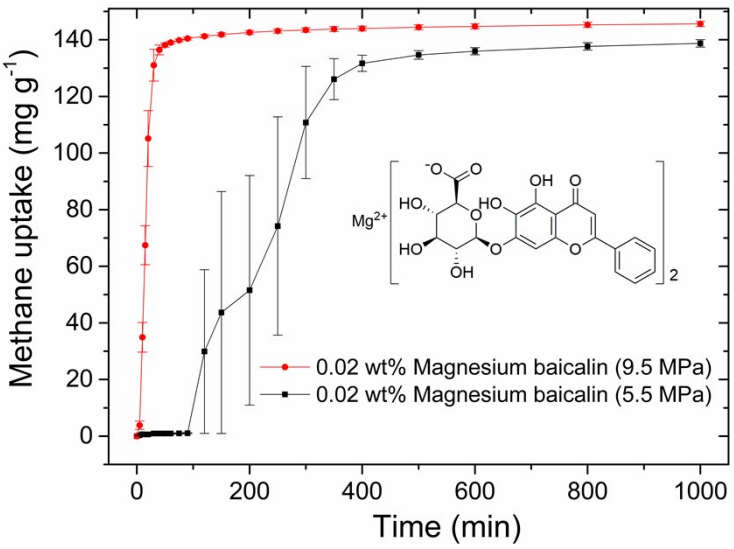

Fig. S7 Methane uptake kinetics for 0.02 wt% magnesium baicalin aqueous solutions at 273 K (initial pressure: 5.5 MPa *versus* 9.5 MPa).

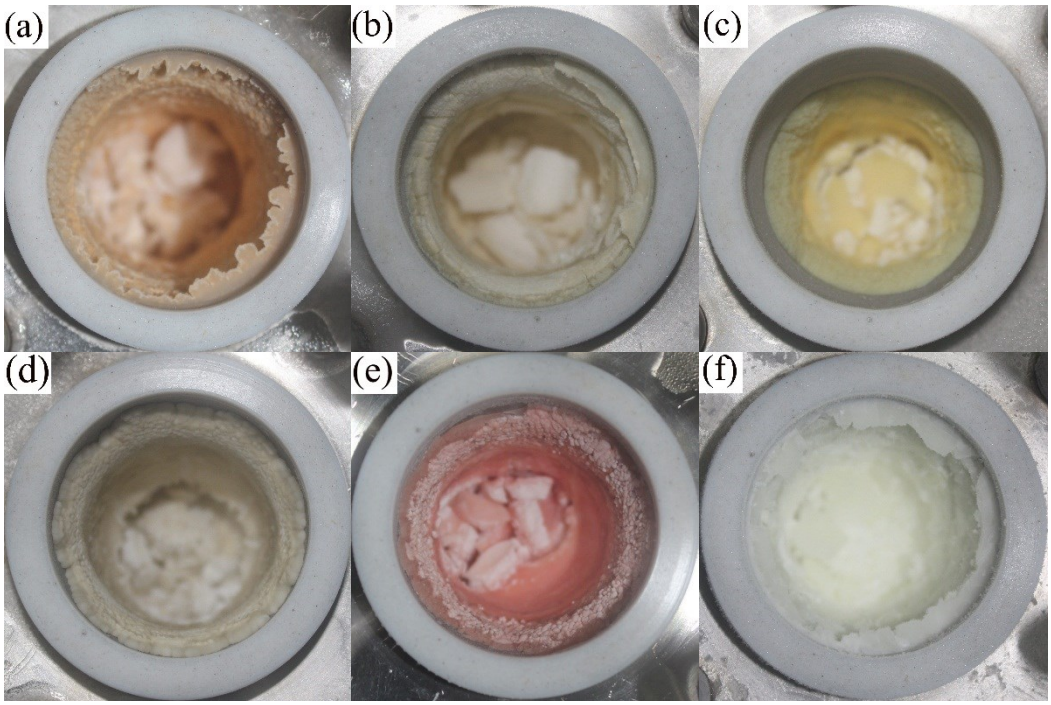

Fig. S8 Methane hydrate grew upward on the vessel wall. (a) *Sanguisorbae Radix*; (b) *Chebulae Fructus*; (c) *Scutellariae Radix*; (d) *Rhapontici Radix*; (e) *Rosae Rugosae Flos*; (f) 0.02 wt% magnesium baicalin.

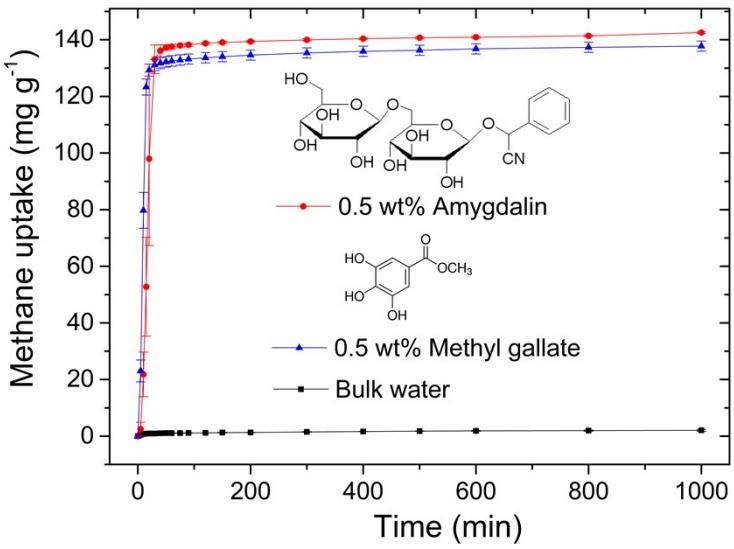

Fig. S9 Methane uptake kinetics for the bulk water, the aqueous solutions of amygdalin and methyl gallate at 273 K (initial pressure, 9.5 MPa).

References

1 Y. Liu, B. Chen, Y. Chen, S. Zhang, W. Guo, Y. Cai, B. Tan and W. Wang, *Energy Technol.*, 2015, **3**, 815-819.

2 W. Wang, C. L. Bray, D. J. Adams and A. I. Cooper, *J. Am. Chem. Soc.*, 2008, **130**, 11608-11609.