

不同沉积相泥页岩天然气吸附能力 及影响因素

李阳阳¹, 罗良¹, 张民康², 刘佳润¹

1 中国石油大学(北京)地球科学学院; 2 中国石油大学(北京)石油工程学院

摘要 海相、海陆过渡相、陆相泥页岩具有不同的地质特征,导致其天然气吸附能力存在差异。以海相、海陆过渡相、陆相泥页岩等温吸附实验数据与地球化学参数及孔隙结构特征为基础,对比分析不同沉积相泥页岩天然气吸附能力大小及其影响因素。研究表明:泥页岩吸附能力的沉积相顺序为海相>陆相>海陆过渡相。有机质含量、干酪根类型、热演化程度、孔隙结构、矿物组分、温度、压力和湿度对吸附能力均具有影响。有机质与黏土矿物是吸附气的载体,其中有机质是主要吸附载体,有机质含量越高,则吸附气含量越高。成熟度对泥页岩吸附能力的影响表现为“两段式”模式:当成熟度处于临界值以下时,随成熟度增加吸附能力增大;超过临界值,则随热演化程度增加吸附能力减小。有机质类型决定有机质孔的发育潜力,热演化程度通过控制生排烃影响有机质孔的发育程度,三种类型沉积相泥页岩有机质发育类型及成熟程度差异明显,是造成其吸附能力差异的重要原因。温度和压力通过改变吸附-解吸平衡影响泥页岩吸附性;水分子通过占据更多的空间而减小可吸附的气量。

关键词 泥页岩; 吸附能力; 影响因素; 孔隙结构; 海相; 海陆过渡相; 陆相

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0 前言

2000年以来,页岩油气已逐渐成为全球油气勘探开发的重要方向之一。中国具有丰富的页岩油气资源,页岩层系油气也是当前勘探开发的热点领域,已相继建成多个国家页岩气示范区。随着储层压裂改造技术的进一步发展、勘探力量与资金的进一步投入,中国页岩气资源的开采必将迎来蓬勃发展的局面。

页岩气是主要以吸附或游离方式存在于泥页岩中的天然气,具有成因类型多样、储集空间致密的特征,且其源岩层和储层为同一套富有机质泥页岩层系,表现为典型的自生自储成藏模式^[1]。游离气是存在于孔隙和裂缝空间内的天然气;吸附于有机质和黏土颗粒表面的为吸附气;溶解气则溶解于水、沥青质和液态原油中^[2]。泥页岩中溶解气占比相对较低,主要以游离气和吸附气形式存在,学者也往往将二者的含量总和作为总含气量^[3]。泥页岩含气量的确定是进行资源量精确评估和勘探开

发工作的必要步骤,主要通过现场解吸测试和等温吸附实验两种手段获得,需要注意的是后者得到的吸附气含量是泥页岩理论的极限值,一般大于其实测值。

诸多学者已针对泥页岩吸附能力的影响因素进行了大量研究和实验分析。现有的研究大多从陆相、海相等单一沉积相角度分析泥页岩吸附能力特征及其影响因素。然而,不同沉积环境下形成的泥页岩发育特征具有差别,其对应的吸附气体能力也应有所差异。本文统计、总结了国内外关于不同沉积相泥页岩的等温吸附实验数据及相关的地球化学、孔隙结构特征,并进行数据相关度对比,明确了不同沉积相泥页岩吸附能力及其主控因素。

1 不同沉积相泥页岩发育特征及天然气吸附能力

按照泥页岩发育环境与特征,可将其分为海相、海陆过渡相(简称过渡相)和陆相。寒武纪至第四纪,富有机质泥页岩在3种环境下均有发育,且发

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第一作者: 李阳阳, 中国石油大学(北京)本科在读,研究方向为页岩油气储层评价。通信地址:102249北京市昌平区府学路18号
中国石油大学(北京)地球科学学院; E-mail: 372620852@qq.com

育形成了十余套富有机质泥页岩层系^[4]。海相泥页岩多形成于台盆或深水陆棚环境,主要发育于南方扬子区震旦系陡山沱组、寒武系筇竹寺组、奥陶系大乘寺组、奥陶系五峰组—志留系龙马溪组,塔里木盆地寒武系玉尔吐斯组,以及湘中地区泥盆系—石炭系等层系,具有分布面积大、单层厚度大、横向变化小、硅质富集等特点;过渡相泥页岩沉积环境多为三角洲与潟湖,主要发育于鄂尔多斯、四川、渤海湾、湘中、南华北等盆地的石炭系—二叠系,单层较薄,但层数多,常与煤层、致密砂岩互层发育;陆相泥页岩主要形成于湖泊环境,普遍发育于中生代地层,如鄂尔多斯盆地三叠系延长组、四川盆地侏罗系、松辽盆地白垩系青山口组、渤海湾盆地古近系沙河街组和孔店组、江汉盆地古近系潜江组等层系,分布面积较小,累计厚度大,受构造运动影响弱^[1,4-6]。

由表1的统计可知,国内3种沉积相的泥页岩具有如下特征:①陆相、过渡相泥页岩的 TOC 高,海相泥页岩 TOC 较高。②海相泥页岩成熟度(R_o)普遍高于2%,处于过成熟生气阶段;过渡相泥页岩成熟度较高, R_o 多大于1.6%,处于高成熟—过成熟阶段;陆相泥页岩则成熟度适中,多处于成熟生油时期。③海相泥页岩干酪根主要为Ⅰ—Ⅱ₁型;过渡相泥页岩干酪根为Ⅱ—Ⅲ型;陆相泥页岩干酪根以Ⅰ—Ⅱ型为主。④海相泥页岩脆性矿物较多,其中石英占比超过矿物总组分的40%,黏土矿物占比少于石英;陆相泥页岩中,黏土矿物在矿物总组分中占主要地位;过渡相泥页岩矿物组成介于两者之间。

不同沉积相泥页岩吸附天然气能力存在差异,这种差异体现在其相应的吸附气含量上。当前定量描述吸附气含量主要采用等温吸附实验的方式,用Langmuir吸附模型方程表征其吸附能力。Langmuir等温吸附数学表达式如下:

$$V = \frac{P \cdot V_L}{P_L + P}$$

式中: V 为压力为 P 条件下的等温吸附值, m^3/t ; P 为实验压力,MPa; V_L 为Langmuir体积,即给定温度下的气体极限吸附气量, m^3/t ; P_L 为Langmuir体积达到1/2时对应的压力,MPa。Langmuir模型方程拟合得到的是泥页岩理论上的最大吸附气含量,一般大于

实测值。

对前人^[10,36-43]用等温吸附实验得到的不同沉积相泥页岩吸附气含量进行了统计,统计结果见表2。由表2可见,在单位质量下,海相泥页岩吸附气含量最大,陆相泥页岩次之,过渡相泥页岩的吸附气含量最小,这表明不同沉积相泥页岩吸附能力强弱的顺序为海相>陆相>过渡相。不同沉积相泥页岩具有不同的地质特征,这些地质特征造成了其吸附能力的差异。

2 泥页岩吸附能力影响因素

2.1 有机质丰度对泥页岩吸附能力的影响

学者早期实验发现,当泥页岩中 TOC 高时,其可吸附的天然气更多^[44]。Ross等^[45]对加拿大东北部Gordondale页岩的研究得出相似的结论,即 TOC 与吸附气体的能力存在正相关性。后续众多学者在对泥页岩吸附影响因素的研究中也逐渐形成了有机质是泥页岩吸附能力主要贡献者的共识^[9,42]。

本文统计了大量的海相、过渡相、陆相泥页岩等温吸附实验数据及相关地球化学参数和孔隙结构特征,并据此对比分析不同沉积相泥页岩 TOC 与其对应的等温吸附含气量(V_L)之间的关系。选取的海相泥页岩来自南方扬子区龙马溪组与牛蹄塘组,共75个实验数据;选取的过渡相泥页岩来自四川龙潭组、鄂尔多斯盆地太原组,共54个样品实验数据;选取的陆相泥页岩来自鄂尔多斯盆地延长组、渤海湾盆地沙河街组、库车凹陷三叠系—侏罗系,共62个实验数据(图1)。

统计分析结果表明, TOC 越高,泥页岩吸附能力越强。三种沉积相泥页岩均存在这一规律,而且均具有良好的正相关性。其原因是微孔在有机质中较发育,其亲和性强,对甲烷具有更好的吸附性,因此随有机质丰度的增加,微孔体积和占比增大,泥页岩吸附能力增强^[61]。

同时,三种沉积相泥页岩 V_L 相对于 TOC 增加的幅度有所差异:海相泥页岩的 TOC 与 V_L 线性拟合系数达0.794 3,陆相泥页岩为0.556 0,过渡相泥页岩仅为0.234 7,海相泥页岩拟合系数显著高于陆相与过渡相。这种差异则是其他影响因素共同作用的结果。

表1 不同沉积相泥页岩地质特征(数据引自文献[7~35])
Table 1 Geological characteristics of shale of different sedimentary facies (data from references [7~35])

沉积相	层位	盆地/地区	区块/钻井	TOC/%		R_o /%		干酪根类型	孔隙度/%		石英含量/%		黏土矿物含量/%		来源文献
				范围值	平均值	范围值	平均值		范围值	平均值	范围值	平均值	范围值	平均值	
海相	龙马溪组 (S ₁ l)	四川盆地	焦石坝	0.55~5.89	2.54	2.20~3.06		I	1.17~7.22	4.52	50.0~80.0	37.3	16.6~62.8	40.5	[7]
			长宁	2.70~3.25		2.10~3.70		I	2.78~6.12		50.0~80.0		16.6~62.8	30.5	[8]
			威远	0.17~4.30	2.39	2.40~3.00		I、II ₁	3.00~6.00	4.20			13.0~59.0		[9]
			威远	2.10~5.90	3.50	2.00~3.00	2.70	I、II ₁	3.60~5.20	4.30					[10]
			昭通	0.60~6.50	2.00	2.80~3.00		I、II ₁	1.20~7.20	4.50				33.1	[11]
			彭水	1.02~2.29	1.51	1.93~2.26	2.03						18.5~53.2		[12]
	渝东南		彭水	2.60~4.20	3.20	2.40~3.10	2.80		1.80~4.50	2.67					[10]
				0.22~5.31	1.78	1.78~2.93	2.38								
		湘西北	永页1井	0.98~3.47	2.00	2.36~2.48	2.40		1.60~2.00	1.70	37.0~66.0	50.2	12.1~32.0	24.2	[13]
			花页1井	1.27~9.94	2.40	2.00~3.60	2.80				21.4~70.4	43.0	8.9~45.3	31.3	[14]
	湘西北		慈页1井	1.61~3.22	2.28	1.52~2.04	1.76				27.6~59.1	41.6	16.9~42.9	27.2	[15]
			威201井	2.00~3.60	2.80	3.20~3.60	3.30				29.0~72.5	38.5			
海相	牛蹄塘组 (E ₁ n)	湘西北	宜地2井	0.52~5.96	2.05	2.18~2.30	2.26				5.1~55.7	25.6	2.7~61.3	33.3	[16]
			永凤1井	1.13~7.91	5.97	1.76~2.31	1.99	I			5.9~60.6	42.3		20.5	[17]
		黔北		0.70~15.75	3.52	3.50~4.20	3.58	I、II ₁	0.10~6.40	1.48			3.0~47.2	19.6	[18]
				0.22~22.15	4.73	1.60~4.00		I	0.40~17.60	4.64	31.0~77.0	58.3			[19]
		贵州		0.85~35.70	6.73	1.95~2.40	2.22	III	2.00~8.00	6.30	4.2~46.2	20.1	20.3~92.3	61.9	[20]
			东页深1井	0.57~18.37	3.23	1.96~2.40	2.22		1.13~9.00	5.53	0.3~71.9	22.1	6.2~90.6	48.3	[21]
	龙潭组 (P ₃ l)	湘中坳陷		0.06~8.70	2.63	1.11~1.39	1.31	II ₂	0.54~13.82	2.60	7.0~69.0	34.4	11.0~56.0	24.0	[22]
			西页1井	0.40~17.85	4.25	2.68~3.48	3.06	III	1.89~2.83	2.26		47.8		41.4	[23]
		四川盆地	华蓥山	0.15~26.60	3.16	1.09~1.39	1.20	III			12.0~38.0	23.52	66.24		[24]
				0.36~14.56	4.48	2.12~3.52	2.59	II	0.40~1.00		9.0~61.0	33.0	5.0~68.0	38.0	[25]
海陆过渡相	太原组 (P ₁ l)	涟源凹陷		0.70~8.10	2.35	1.67~1.98	1.76	II ₁ 、II ₂							[26]
			临兴	0.37~12.01	3.65	0.97~3.19	1.39	III							[27]
		鄂尔多斯盆地	绥德	0.75~5.71	2.75	1.43~2.12	1.80				2.0~92.4	44.4	3.1~98.0	49.1	[28]
				0.12~22.78	>2.5	1.72~2.92	2.32	III	0.63~3.99	1.66			6.27~65.87		[29]
	本溪组(C ₂ b) 山西组(P ₁ sh)	鄂尔多斯盆地	延长探区	0.50~3.00	0.81			III	3.00~8.00		35.0~56.0	45.8	43.0~53.0	47.0	[30]
				1.00~11.00	3.73			III	2.00~5.00		6.0~77.0	38.7	23.0~87.0	58.0	
	延长组 (T ₁ y)	鄂尔多斯盆地	延长探区	2.73~8.82	4.93	0.83~1.10		II					24.7~59.7	42.8	[31]
			甘泉	0.46~25.46		1.25~1.33		II ₁	0.50~4.00	1.82	20.0~30.0	26.3	37.4~72.8	40.0	[32]
海相	沙河街组 (E ₁₋₂ sh)	渤海湾盆地		1.76~6.28	3.92	0.50~1.20		I、II	1.69~6.83	3.83		27.75		42.11	[33]
			辽河坳陷	0.50~7.41	2.46	0.50~1.32		II ₁	2.10~8.20	3.86			9.95~53.0	41.85	[34]
			济阳坳陷	2.00~10.50		0.50~1.40		I、II ₁	3.00~16.0						[35]

表2 不同沉积相泥页岩吸附气含量(数据引自文献[10,36-43])
Table 2 Adsorbed gas content of shale of different sedimentary facies(data from references [10,36-43])

沉积相	层位	盆地/地区	区块	吸附气含量/($\text{m}^3 \cdot \text{t}^{-1}$)		来源 文献
				范围值	平均值	
海相	龙马溪组(S_1l)	四川盆地	焦石坝	3.8~9.6	6.80	[36]
			威远	3.8~7.3	5.80	
			长宁	3.9~6.5	4.90	[10]
			昭通	2.2~6.5	3.90	
海陆过渡相	牛蹄塘组(E_1n)	黔北	岑巩	1.45~6.09	3.78	[37]
	太原组(P_1t)	南华北	中牟	1.03~3.13	1.74	[37]
	龙潭组(P_3l)	黔北		1.51~3.16	2.33	[38]
		下扬子		1.70~4.82	2.81	[39]
	山西组(P_1sh)	鄂尔多斯盆地		0.96~2.71	1.94	[40]
陆相	延长组(T_3y)	鄂尔多斯盆地	下寺湾	2.90~4.56	3.37	[41]
			志丹—甘泉	0.56~4.43	2.26	[42]
	青山口组(K_2qn)	松辽盆地		1.71~3.61	2.97	[43]

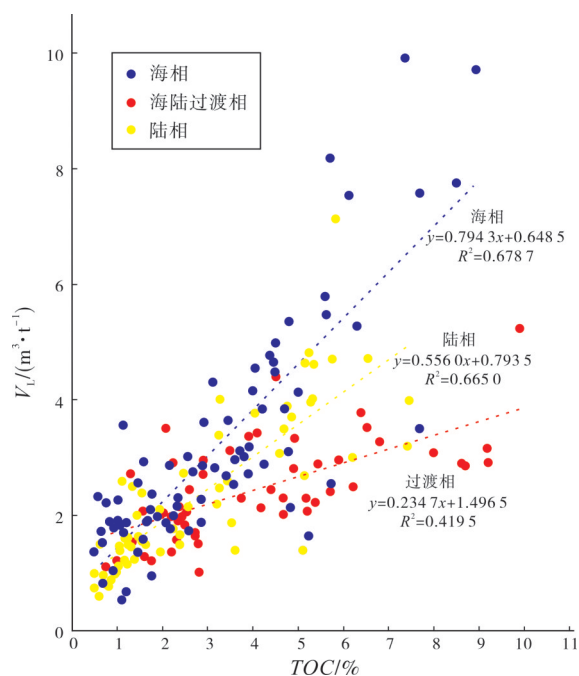


图1 三种沉积相泥页岩TOC与 V_L 相关关系
(数据引自文献[29,34, 38,46-60])

Fig. 1 Correlation between TOC and V_L of shale of three types of sedimentary facies (data from references [29,34, 38,46-60])

2.2 有机质成熟度对泥页岩吸附能力的影响

泥页岩吸附能力也受有机质成熟度的影响。目前关于其对泥页岩吸附能力影响的报道结论不一,主要有以下3种情况:

(1)随有机质热演化程度的增加,泥页岩的吸附能力呈增大趋势^[62]。该情况主要是由于随有机质热演化的进行,泥页岩生烃产生大量的有机质

孔,使孔隙类型由以宏孔及中孔为主向以纳米级有机质孔为主变化,这一变化有助于增大有机质的比表面积,并且有机质生烃的同时会产生有机酸,可导致形成粒内溶蚀孔,为泥页岩提供更多的空间来吸附页岩气,从而提高了可吸附的气体量^[37,63]。

(2)成熟度对泥页岩吸附能力的影响表现为“两段式”,最大吸附气量(V_L)与泥页岩成熟度之间表现出先增后降的趋势:即有临界点 R_0 值,当成熟度处于临界值以下时,随成熟度增加吸附能力增大;超过临界值,则随热演化程度增加吸附能力减小。 R_0 超过临界值,吸附能力呈现下降趋势的原因:一是有机质成熟度的增加导致的TOC下降的不利影响超过有机质孔发育的有利影响^[34];二是泥页岩埋深过大(>3 000 m)时,压实作用强烈,使有机质孔发生坍塌或被充填,造成有效连通孔隙减少,从而使吸附能力降低^[64]。

(3)随热演化程度的增加泥页岩可吸附的气体呈减小趋势。Chalmers等^[65]在对西加拿大盆地白垩统Buckingham组页岩进行研究后,发现泥页岩吸附能力与热演化程度呈负相关性。

尽管不同学者对于热演化程度影响泥页岩吸附能力的研究结果具有差异,但对成熟度通过控制有机质孔隙发育而影响泥页岩吸附性的观点却比较一致。对前人大量研究的进一步梳理发现,上述3种情况可归纳于同一种模式,即“两段式”模式,正相关及负相关的情况应该是先升后降其中一段的反映。先升后降的“两段式”中,不同沉积相泥页岩的临界值

相差较大。本文对保靖地区龙马溪组页岩的吸附气含量与热演化的相关性进行分析后发现,其 R_o 临界值为3.36%^[53](图2a)。高和群等^[62]对渝东南彭水地区龙马溪组的研究显示成熟度与吸附能力呈正相关,其原因应该是所研究的泥页岩样品 R_o 未达到临界点(图2b),故与“两段式”模式并不矛盾。对过渡相而言,本文对黔北地区龙潭组泥页岩的分析发现 R_o 临界值为2.58%^[38](图2c);熊荃等^[29]对鄂尔多斯盆

地太原组泥页岩的研究显示, R_o 临界值为1.7%左右(图2d)。陆相泥页岩中,对川西地区须家河组页岩、库车坳陷侏罗系泥页岩的分析发现,前者 R_o 临界值为1.3%^[62](图2e),后者 R_o 临界值在1.0%左右^[64](图2f)。可见在三种沉积相泥页岩中,海相泥页岩 R_o 临界值最高,过渡相泥页岩次之,陆相泥页岩较低。所以 V_L 与 R_o 的正相关多见于海相泥页岩,而陆相泥页岩中多有“两段式”及负相关的现象出现。

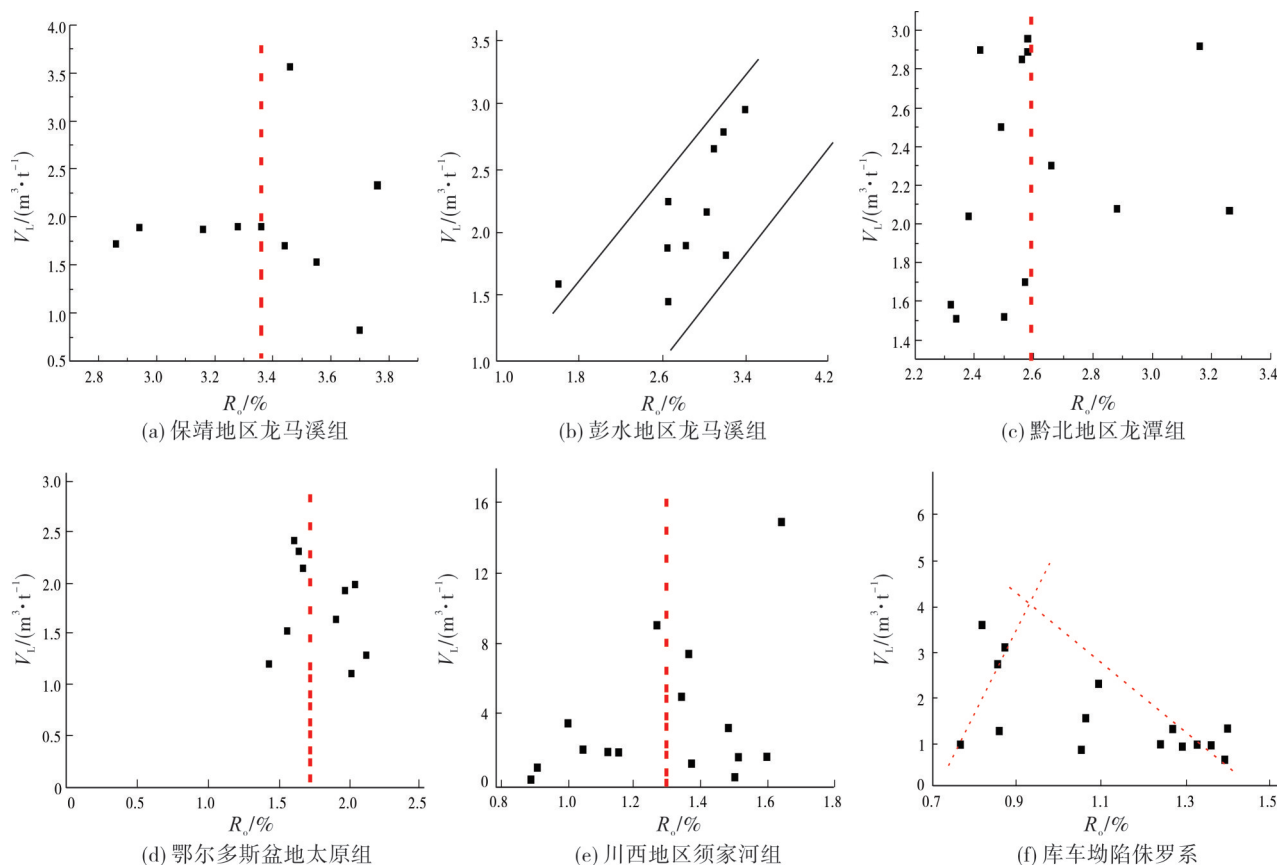


图2 不同地区泥页岩 R_o 与 V_L 关系 (a据文献[53]修改;b据文献[62];c据文献[38];d据文献[29]修改;e据文献[62]修改;f据文献[64]修改)

Fig. 2 Relationship between R_o and V_L of shale in different areas(cited from references [29,38,53,62,64])

2.3 有机质类型对泥页岩吸附能力的影响

有机质类型是指干酪根类型,根据主要元素的组成可划分成Ⅰ型、Ⅱ型和Ⅲ型,其对泥页岩吸附能力也具有较大的影响。海相泥页岩有机质以Ⅰ型—Ⅱ₁型为主;过渡相泥页岩多发育Ⅱ型和Ⅲ型有机质;陆相泥页岩有机质类型比较复杂,深湖相以Ⅰ型为主,浅湖相多发育Ⅱ型,湖沼相多发育Ⅲ型,总体呈现以Ⅱ型为主、兼有Ⅰ型与Ⅲ型^[1,41]。

有机质类型对泥页岩吸附能力的影响主要有

两个方面:一是不同类型的有机质本身的吸附能力不同;二是在有机质热演化过程中,热演化程度对不同类型有机质吸附能力的影响不同。

对于前者,薛冰等^[66]通过统计发现不同类型泥页岩的等温吸附气含量范围和单位有机质吸附气含量都存在差异,并且有机质吸附能力表现为Ⅲ型>Ⅱ型>Ⅰ型。其原因在于不同有机质类型的显微组分与微观结构不同,相对于Ⅰ型而言,Ⅲ型有机质具有更多的芳香结构和更高的镜质组含量,而镜质组、芳环结构对甲烷表现出更强的亲和力^[67]。

对于后者,侯宇光等^[68]研究发现,Ⅲ型有机质在热演化过程中几乎不发育有机质孔隙;陈中红等^[69]研究发现,相对于Ⅲ型有机质,Ⅰ型有机质在热演化过程中更有利于发育有机质孔。而有机质孔的发育可提供更多的吸附位点,从而明显提高可吸附的含气量。

虽然Ⅲ型有机质本身的吸附能力更强,但其生烃能力和发育有机质孔的潜能差,而Ⅰ型、Ⅱ型有机质生烃潜力大,易发育有机质孔。综合来看,有机质随热演化生成有机质孔对吸附能力的影响比有机质本身吸附能力的影响更大,故有机质类型对吸附性影响的顺序应为Ⅰ型>Ⅱ型>Ⅲ型。

2.4 矿物组分对泥页岩吸附能力的影响

泥页岩中的矿物由黏土矿物和脆性矿物2类组成。陆相、过渡相泥页岩,其黏土矿物占比普遍高于海相泥页岩(图3),而脆性矿物含量则刚好相反,海相泥页岩普遍具有较高含量的石英,陆相、过渡

相泥页岩石英含量相对较低(图4)。

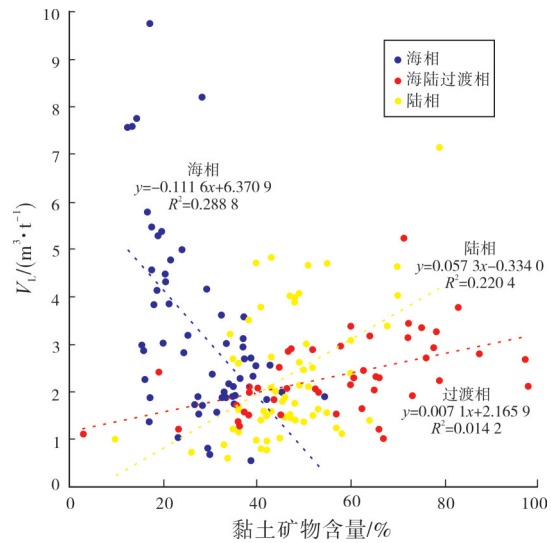
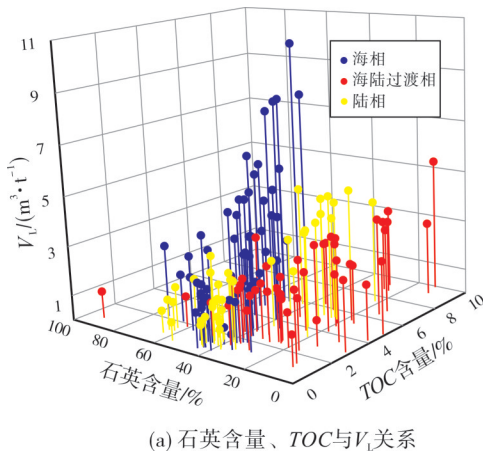
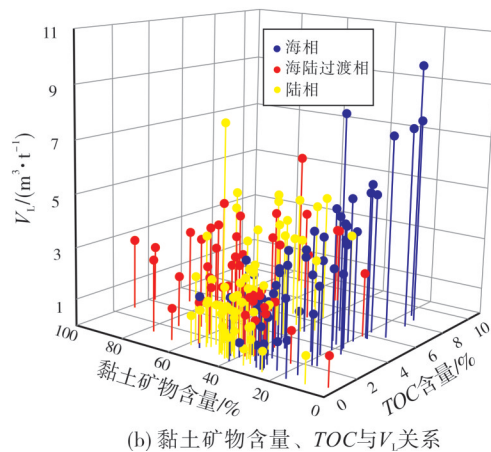


图3 三种沉积相泥页岩黏土矿物含量与 V_L 相关关系
(数据引自文献[29,34,38,46-60])

Fig. 3 Correlation between clay mineral content and V_L of shale of three types of sedimentary facies (data from references [29,34,38,46-60])



(a) 石英含量、TOC与 V_L 关系



(b) 黏土矿物含量、TOC与 V_L 关系

图4 三种沉积相泥页岩 V_L 与矿物组分关系(数据引自文献[29,34,38,46-60])

Fig. 4 Correlation between mineral composition and V_L of shale of three types of sedimentary facies (data from references [29,34,38,46-60])

海相泥页岩等温吸附 V_L 与黏土矿物含量为负相关性,与石英含量为正相关性,而海陆过渡相、陆相泥页岩 V_L 则与黏土矿物含量具有良好的正相关,与石英含量具有负相关(图3,图4)。矿物成分对泥页岩吸附能力的影响出现这种相反现象的原因是:海相泥页岩沉积环境不利于陆源碎屑黏土的大量搬运与沉积,而石英多为生物成因,常伴随有机质富集,因此出现石英含量随TOC增加而增加的现象,相对的黏土矿物含量占比则会变小。根据前文可知,海相泥页岩 V_L 与TOC存在显著正相关性,故在海相泥页岩中,出现了 V_L 与石英含量呈正相关的

现象,而实际上石英对吸附能力的影响很小。

陆相泥页岩中的石英主要来源于陆源碎屑的搬运,其含量受物源距离的影响:距物源越远,搬运能力越弱,石英沉积就越少,相对的黏土矿物占比就越大^[41]。同时,在陆相和海陆过渡相环境,黏土矿物及其发育环境有利于有机质的生产和保存。黏土矿物对于泥页岩吸附的影响是通过其比表面来实现的。吴陈君等^[70]对比不同黏土矿物含量的泥页岩孔隙结构特征,发现富含黏土矿物的泥页岩具有更高的比表面积与吸附量。不同黏土矿物由于自身粒径与晶体结构的差异,其对吸附性的影响

也有不同:在相同的压力下,黏土矿物吸附能力顺序为蒙脱石>伊-蒙混层>高岭石>绿泥石>伊利石^[71](图5)。

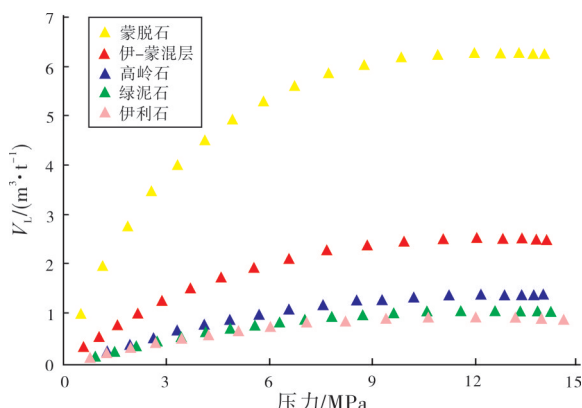


图5 不同类型黏土矿物的吸附能力(据文献[71]修改)

Fig. 5 Adsorption capacity of different types of clay minerals
(cited from reference reference [71], modified)

2.5 孔隙结构对泥页岩吸附能力的影响

孔隙结构是影响泥页岩吸附性的最主要因素。孔隙结构特征包括孔隙比表面积、总孔体积、平均孔隙直径等,其中比表面积对吸附性的影响更为关键。有机质是泥页岩赋存页岩气的主要载体,吸附气的主要赋存空间为有机质表面。前文所述的吸附能力的其他影响因素,都可归结于通过控制孔隙的比表面积来实现对泥页岩吸附的影响^[72]。三种沉积相泥页岩的比表面积与 V_L 均呈现较好的正相关(图6)。

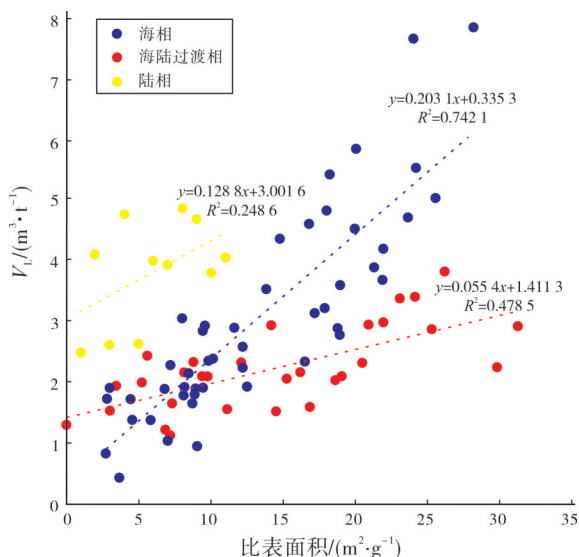


图6 三种沉积相泥页岩 V_L 与比表面积相关关系

(数据引自文献[29,34,38,46-60])

Fig. 6 Correlation between specific surface area and V_L of shale of three types of sedimentary facies (data from references [29,34,38,46-60])

孔隙直径对比表面积有重要影响。按照孔隙直径的大小,可将孔隙划分为微孔、中孔和大孔。钟玲文等^[73]研究发现微孔发育的泥页岩,其比表面积更大,相对中孔和大孔,小孔对泥页岩的吸附具有更大的贡献。冯小龙等^[74]对鄂尔多斯盆地延长组泥页岩的实验表明:中孔体积占总孔体积的76.69%,贡献的比表面积却只有67.84%;微孔体积占比为10.12%,贡献的比表面积却达到了30.27%。杨潇等^[75]在对渝东南牛蹄塘组、龙马溪组泥页岩孔隙结构的研究中发现:微孔体积占比为20%,对比表面积的贡献达57.6%;中孔体积占比为64%,对比表面积的贡献为42.35%。这些均表明微孔对泥页岩吸附的影响更大。

2.6 温度和压力对泥页岩吸附能力的影响

温度和压力对泥页岩吸附也有影响^[76]。前人研究发现泥页岩 V_L 与温度具有负相关性,其随温度的升高而下降^[44](图7a)。而压力对 V_L 的影响则不同,表现为正相关性:随着压力的增大, V_L 的增加先快后慢,最后趋于平稳(图7b)。

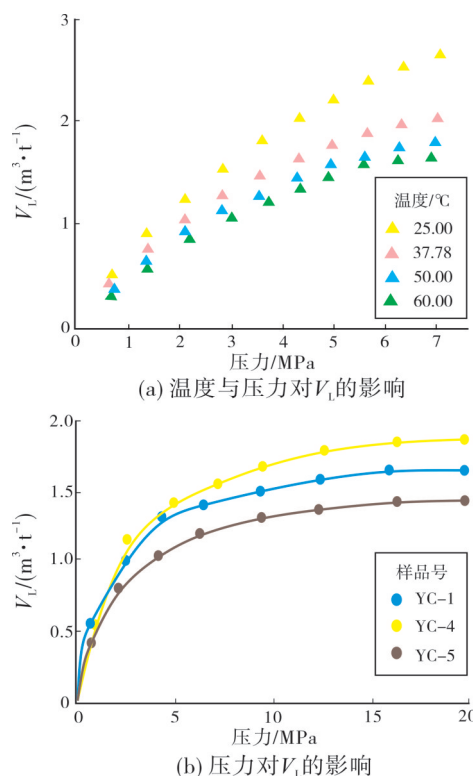


图7 温度与压力对泥页岩 V_L 的影响

(图a据文献[44]修改,图b据文献[77]修改)

Fig. 7 Influence of temperature and pressure on V_L of shale
(Fig.a modified from reference [44], Fig.b modified from reference [77])

2.7 湿度对泥页岩吸附能力的影响

湿度对泥页岩吸附也有影响。张烈辉等^[78]对川南龙马溪组泥页岩样品进行了干燥与平衡湿度的对比实验(图8),结果表明:水分会制约泥页岩的吸附性,且随含水率的增加,泥页岩 V_L 逐渐减小。Hartman^[79]对 Barnett 页岩的研究也得到相同结论。其原因是水分子的极性高,更易与有机质或黏土矿物发生吸附,占据了部分可供吸附的空间,使得气体可占据的有效吸附面积变小,同时水分的存在会阻碍气体在孔隙中的渗滤和扩散作用,从而减少了吸附气含量。

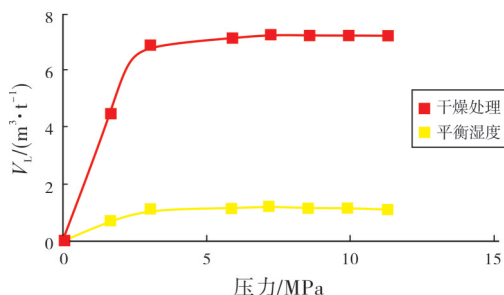


图8 湿度对泥页岩 V_L 的影响(据文献[78]修改)

Fig. 8 Influence of humidity on V_L of shale (cited from [78], modified)

3 讨论

3.1 不同沉积相泥页岩吸附能力的差异性

不同沉积相泥页岩的吸附能力存在明显差异(表2)。总体而言,海相泥页岩吸附能力高于陆相,过渡相泥页岩的吸附能力较弱,即泥页岩吸附能力的沉积相顺序表现为海相>陆相>过渡相。泥页岩吸附能力表现的是前文所述7个因素综合影响的结果。

海相泥页岩有机质类型为Ⅰ型—Ⅱ₁型,发育有机质孔潜力大,随着生烃过程的进行,发育大量的有机质孔,孔隙类型向以纳米级有机质孔为主变化,该变化有助于提高吸附能力。我国海相泥页岩的有机质成熟度普遍较高,这保证了其能够发育大量的有机质孔隙。热演化对吸附能力的影响不是单一的,热演化一方面促进有机质孔的发育,一方面降低有机质含量。海相泥页岩 TOC 虽低于陆相、过渡相,但其有机质类型有利于有机质孔的形成,故初始热演化进程的有利影响占主导地位,并一直持续到很高成熟度后才停止。从图6可知,海相泥页岩具有较大的比表面积,也印证了这一点。海相

泥页岩的高成熟度及有机质类型决定了其吸附能力最大。虽然陆相泥页岩同海相一样具有利的有机质类型,但热演化对其的影响却不如海相泥页岩。过渡相泥页岩有机质以Ⅱ型—Ⅲ型为主,Ⅲ型干酪根在热演化过程中几乎不发育有机质孔。同时,陆相相对于过渡相具有更高占比的黏土矿物含量(图3),而黏土矿物对泥页岩的吸附起到积极作用,所以陆相泥页岩的吸附能力大于过渡相。

3.2 同一沉积相泥页岩吸附能力的差异性

不同沉积相泥页岩吸附能力存在差异,但同一沉积相之间也存在差异。以海相为例,南方地区寒武系筇竹寺组与志留系龙马溪组同为海相,但在地质特征与吸附能力上具有明显差异。前者热演化程度过高,有机质孔不发育,孔隙度相对较低,泥页岩含气量低;后者热演化程度适中,有机质孔隙发育,泥页岩含气量高。赵文智等^[10]对长宁—威远地区筇竹寺组与龙马溪组的研究发现,前者页岩含气量仅达到后者的1/2左右。同时,南方地区龙马溪组、筇竹寺组富有机质页岩集中段钻井测试产量显示,前者产气量较好,后者为无气—微气^[10]。这种差异也存在于陆相、过渡相泥页岩中。究其原因,在于海相、陆相等的概念过于宏观,均涵盖了较多的沉积相、亚相及微相,不同研究区具体的沉积环境、构造运动、保存条件、热演化史等地质条件都具有较大的差异,因此泥页岩的吸附能力还需要在普遍性规律的基础上进行具体分析。

3.3 存在的问题及研究方向

在测定泥页岩孔隙结构时,大多数学者采用氮气等温吸附实验,而氮气等温吸附实验主要适用于中孔范围内的孔隙直径与比表面积测定,对于微孔发育的海相泥页岩来说,会存在较大误差。将氮气等温吸附实验、CO₂吸附实验、高压压汞实验3种方法相结合,测定精度与实验严谨性会得到很大提升。

孔隙结构从根本上决定泥页岩的吸附能力,热演化进程对于孔隙结构也有着重要影响。已有的关于成熟度对于泥页岩吸附影响的研究结果存在多种情况,本文梳理认为多种情况可归纳于同一种模式,即“两段式”模式,但是否具有普遍性与代表性仍需要进一步的验证。可选取海相、陆相、过渡

相典型层位的具有不同成熟度、不同有机质类型的系列泥页岩,进行全面的油气地球化学实验,获得有机地球化学特征;采用氮气吸附、CO₂吸附及高压压汞相结合的方式精准测定孔隙参数特征;通过氩离子抛光—扫描电镜,获得其微观结构特征,进而进行综合分析研究,一方面可以检验“两段式”模式是否具有普遍性,另一方面可以研究热演化进程中不同沉积相泥页岩的孔隙发育特征,这将有助于深化不同沉积相泥页岩天然气吸附能力及影响因素的认识。

4 认识与结论

有机质含量、有机质类型、热演化程度、孔隙结构、矿物组分、温度、压力和湿度对泥页岩吸附能力均具有影响。有机质与黏土矿物是吸附页岩气的载体,其中有机质是主要吸附载体,通常情况下,有机质含量高,则页岩气含量高。孔隙比表面是吸附气的主要赋存空间,也是从根本上决定不同沉积相泥页岩吸附能力的因素。有机质类型决定有机质孔的发育潜力,热演化程度通过控制生排烃过程来影响有机质孔的发育程度;温度和压力通过改变吸附—解吸平衡影响泥页岩吸附性;水分子通过占据更多的空间而减小可吸附的气量。

不同沉积相泥页岩发育类型及吸附能力存在差异,总体而言,泥页岩吸附天然气能力的沉积相顺序为海相>陆相>过渡相。相同沉积相之间也存在差异,由于海相、海陆过渡相、陆相的概念过于宏观,均涵盖了较多的沉积相、亚相及微相,且不同地区的地质条件也具有较大差异,这导致了同一类型相区泥页岩吸附能力的差异,因此实际工作中还需要针对具体研究区域结合地质条件来做进一步的对比和分析。

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Gas-adsorption capacity of shale of different sedimentary facies and its influencing factors

LI Yangyang, LUO Liang, ZHANG Minkang, LIU Jiarun

Abstract: Shale of different sedimentary facies has different geological characteristics, which leads to different gas-adsorption capacity. Based on the isothermal adsorption experimental data, geochemical parameters and pore structure characteristics shale of marine, marine-continental transitional and continental, the gas-adsorption capacity and influencing factors of shale of different sedimentary facies are compared and analyzed. In general, the order of shale gas-adsorption capacity is marine facies > continental facies > marine-continental transitional facies. Organic matter content, kerogen type, thermal evolution degree, pore structure, mineral composition, temperature, pressure and humidity all affect the adsorption capacity. Organic matter and clay minerals are the carriers of adsorbed gas, in which organic matter is the main adsorption carrier, so the higher the content of organic matter, the higher the content of adsorbed gas. The influence of maturity on the adsorption capacity of shale shows a "two-stage" mode: when the maturity is below the critical value, the adsorption capacity increases with the increase of maturity; above the critical value, the adsorption capacity decreases with the increase of maturity. The type of organic matter determines the development potential of organic matter pores, and the degree of thermal evolution affects the development degree of organic matter pores by controlling hydrocarbon generation and expulsion. There are obvious differences in the type and maturity of organic matter in shale of the three types of sedimentary facies, which is an important reason for the difference of their adsorption capacity. Temperature and pressure affect the adsorption of shale by changing the adsorption-desorption equilibrium. Water molecules reduce the amount of adsorbable gas by occupying more space.

Key words: shale; gas-adsorption capacity; influencing factor; pore structure; marine facies; marine-continental transitional facies; continental facies

LI Yangyang, First author; Undergraduate of China University of Petroleum(Beijing), engaged in shale reservoir evaluation. Add: School of Geosciences, China University of Petroleum(Beijing), No.18 Fuxue Rd., Changping District, Beijing 102249, China