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东营凹陷古近系泥页岩中可溶有机质特征与页岩油“甜点”预测

何晋译[1],蔡进功[1],雷天柱[2],张守鹏[3],张存霞[3]

(1.同济大学 海洋地质国家重点实验室,上海 200092; 2.中国科学院地质与地球物理研究所 兰州油气资源研究中心,甘肃 兰州 730000; 3.中国石化胜利油田分公司 勘探开发研究院,山东 东营 257015)

摘要:可溶有机质作为页岩油的重要载体,所蕴含的丰富信息对页岩油“甜点”预测具有重要意义。选取东营凹陷沙四段下亚段、沙四段上亚段、沙三段下亚段和沙三段中亚段泥页岩样品,对其可溶有机质族组分及饱和烃色谱进行分析,据可溶有机质含量、族组分及轻质烃含量等特征,确立反映页岩油丰富程度的参数(可溶有机质含量和饱和烃含量)及表征页岩油流动性的参数(流动性指数、轻质烃含量和轻质烃散失量)。结果表明,沙四段上亚段和沙三段下亚段可溶有机质含量与饱和烃含量明显高于沙四段下亚段和沙三段中亚段,以沙四段上亚段最高;沙四段上亚段和沙三段下亚段可溶有机质中流动性指数较沙四段下亚段和沙三段中亚段更高,同时沙四段上亚段具有更高的原始轻质烃含量和轻质烃散失量。综合分析认为,沙四段上亚段和沙三段下亚段是页岩油有利发育层段,其中以沙四段上亚段最好,是东营凹陷古近系页岩油“甜点”优选目标区。

关键词:页岩油;可溶有机质;流动性指数;轻质烃;“甜点”预测

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Characteristics of soluble organic matter of Paleogene shale in Dongying Sag and prediction of shale oil “sweet spots”

HE Jinyi[1], CAI Jingong[1], LEI Tianzhu[2], ZHANG Shoupeng[3], ZHANG Cunxia[3]

(1.*State Key Laboratory of Marine Geology, Tongji University, Shanghai City*, 200092, *China*; 2.*Lanzhou Center for Oil and Gas Resources, Institute of Geology and Geophysics, Chinese Acamedy of Sciences, Lanzhou City, Gansu Province*, 730000, *China*; 3.*Exploration and Development Research Institute, Shengli Oilfield Company, SINOPEC, Dongying City, Shandong Province*, 257015, *China*)

Abstract: As an important carrier of shale oil, soluble organic matter contains abundant information which is of great significance to the prediction of shale oil “sweet spots”. Shale samples drilled from Lower Es_4 Member, Upper Es_4 Member, Lower Es_3 Member and Middle Es_3 Member in Dongying Sag were selected to analyze the group component of soluble organic matter and saturated hydrocarbon chromatography. According to the characteristics of soluble organic matter content, group composition and light hydrocarbon content, indicators including the total amount of soluble organic matter and the amount of saturated hydrocarbon were established to identify the abundance of shale oil, as well as indicators including the *MI* parameter, light hydrocarbon content and loss of light hydrocarbon to characterize the mobility capacity of shale oil. The results indicate that the content of soluble organic matter and saturated hydrocarbon of Upper Es_4 Member and Lower Es_3 Member was significantly larger than that of Lower Es_4 Member and Middle Es_3 Member, and that of Upper Es_4 Member were the highest among the four layers. Shale oil *MI* of Upper Es_4 Member and Lower Es_3 Member was higher than that of

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作者简介:何晋译(1990—),男,山东威海人,在读博士研究生,从事油气赋存机理和石油地质学研究。联系电话:15588302428,E-mail:280282168@qq.com。
通信作者:蔡进功(1961—),男,山东长岛人,教授,博导。联系电话:(021)65988829,E-mail:jgcai@tongji.edu.cn。
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Lower Es_4 Member and Middle Es_3 Member. Meanwhile, the original light hydrocarbon content and loss of Upper Es_4 Member were higher. According to the comprehensive analysis, Lower Es_3 Member and Upper Es_4 Member are favorable development formations of shale oil. Further more, Upper Es_4 Member is the preferred target area of Paleogene shale oil “sweet spots” in Dongying Sag.

Key words: shale oil; soluble organic matter; *MI* parameter; light hydrocarbon; “sweet spots” prediction

页岩油是一种典型的非常规油气资源，随着勘探技术及开发能力的提高，页岩油被认为是最有可能成为替代石油天然气的能源，页岩油的勘探开发引起了中外石油地质工作者广泛关注[1-7]。东营凹陷是中国东部陆相盆地的富油凹陷之一[8]，在沙四段上亚段和沙三段下亚段泥页岩中发现了多口工业油气流井[9-12]。前人对东营凹陷页岩油的储层矿物组成与储集空间特征[13-18]、可动性及可动率[11-12,19]、赋存特征与富集规律[20-22]、利用地化参数预测资源有利区[23-24]等方面进行了深入研究，但对可溶有机质的特征及可溶有机质与页岩油“甜点”间的关系等方面研究较少。可溶有机质作为页岩油的重要载体，其丰富程度及可动性对页岩油“甜点”预测具有重要意义。泥页岩中可溶有机质采用有机溶剂抽提获得，主要以游离态或较弱的物理吸附态赋存于泥页岩孔隙中[25]，其组成与原油接近，能较好地解译页岩油的基本特征[26]。页岩油的可动性是页岩油勘探开发过程中需考虑的一个关键问题[27]，有机溶剂抽提得到的可溶有机质是页岩油可动的最重要组分[28]，其族组分及轻质烃含量均会影响页岩油的黏度及流动性[11,29-36]，也是泥页岩层段能否作为页岩油“甜点”的重要判识参数。因此，围绕泥页岩中可溶有机质开展研究，既可揭示页岩油的黏度及流动性等基本特征，也可为页岩油“甜点”预测提供技术支撑，从而提高页岩油的勘探开发效益。

1 实验样品与方法

1.1 实验样品

样品取自东营凹陷沙四段下亚段(Es_4下)、沙四段上亚段(Es_4上)、沙三段下亚段(Es_3下)和沙三段中亚段(Es_3中)埋深为2 954.7~4 500.8 m的泥质烃源岩。岩性以暗色泥页岩为主，有机质含量总体较高，总有机碳含量(*TOC*)为0.43%~8.30%，均值为3.16%，S_1为0.06~12.23 mg/g(均值为2.95 mg/g)，S_2为0.23~53.26 mg/g(均值为13.75 mg/g)，最高热解峰温(T_{max})显示已进入生烃门限，其值为422~448 ℃(均值为438 ℃)(表1)。研究区沙四段和沙三段均发育湖相泥质烃源岩[37-40]，不同沉积时期形成的泥页岩沉积环境存在较大差异，其中沙四段下亚段普遍发育含膏泥岩、含盐泥岩、膏质泥岩和盐质泥岩，为盐湖相沉积[40]；沙四段上亚段又可细分为纯下次亚段(Es_4上纯下)和纯上次亚段(Es_4上纯上)，纯下次亚段发育块状泥岩和石膏质泥岩，为间歇性咸水湖泊沉积；纯上次亚段主要发育深灰色-灰黑色油页

表1 东营凹陷古近系泥页岩样品基本地球化学特征

Table1 Basic geochemical information of shale samples in Dongying Sag

样品编号	井号	层位	埋深/m	S_1/(mg•g^{-1})	S_2/(mg•g^{-1})	T_{max}/℃	*TOC*/%
1	N38	Es_3中	2 954.7	0.06	1.54	438	1.21
2	N38		3 001.8	0.27	5.87	434	1.97
3	N38		3 049.68	0.13	1.86	438	1.12
4	N38		3 186.84	0.81	12.82	436	2.66
5	N38		3 219.68	0.62	8.26	437	2.28
6	N38	Es_3下	3 264.5	1.47	13.47	438	3.02
7	NY1		3 295.38	7.04	53.26	444	7.55
8	NY1		3 307.76	1.76	11.23	439	1.91
9	NY1		3 309.25	1.59	9.85	440	1.74
10	N38		3 339.6	2.79	36.01	439	5.5
11	N38		3 350.3	2.55	18.02	442	3.24
12	N38		3 364.43	2.15	16.94	442	3.08
13	NY1	Es_4上纯上	3 332.24	2.57	11.66	441	2.36
14	NY1		3 346.96	4.15	19.58	441	3.71
15	NY1		3 362.05	3.16	3.89	431	1.82
16	NY1		3 365.04	1.94	10.89	442	2.16
17	NY1		3 388.05	4.74	19.01	439	3.72
18	NY1		3 400.3	5.96	22.17	443	4.03
19	NY1		3 401.63	4.2	12.93	438	2.52
20	NY1		3 416.14	3.58	19.02	445	3.34
21	NY1		3 434.83	7.76	43.06	444	8.3
22	NY1		3 443.31	2.59	12.9	439	2.51
23	NY1		3 449.86	5.29	19.88	433	5.26
24	NY1	Es_4上纯下	3 484.58	4.46	12.51	437	4.32
25	NY1		3 496.31	12.23	11.71	430	5.42
26	F8	Es_4下	3 946.19	0.64	0.65	422	0.76
27	FS2		3 968.5	1.11	1.32	438	1.19
28	FS2		4 295.3	0.23	0.23	439	0.43
29	XLS1		4 376	2.33	1.67	448	5.63
30	FS2		4 500.8	0.3	0.26	426	0.7

岩，属于常年闭流咸水湖泊沉积；沙三段下亚段沉积了一套黑色页岩和油页岩，为半咸湖深湖相沉积；沙三段中亚段主要发育块状泥岩和粉砂岩，是湖盆水体变浅的结果[38,41-43]。

1.2 实验方法与条件

1.2.1 有机溶剂抽提和族组分分离方法

泥页岩样品充分研磨至80目以上，低温（不高于50 ℃）烘干后一直保持干燥。称取一定数量的泥页岩样品，并记录各样品的重量，在索式抽提器中以正戊烷为抽提剂抽提72 h，控制水浴温度约为39 ℃，得到正戊烷抽提液和残渣。以二氯甲烷为抽提剂，在索式抽提器中对残渣抽提72 h，得到二氯甲烷抽提液。然后浓缩二氯甲烷抽提液，沉淀沥青质并用万分之一精度的天平称重。将正戊烷抽提液和沉淀过沥青质的二氯甲烷抽提液过硅胶氧化铝层析柱。所用硅胶用氯仿抽提至无荧光，在电热烘箱中150 ℃温度活化6 h，中性氧化铝在马弗炉中450 ℃温度活化6 h。用正戊烷反复冲洗氧化铝硅胶柱得到饱和烃，用二氯甲烷反复冲洗得到芳烃，用甲醇冲洗得到非烃。将饱和烃、芳烃和非烃转移至已经恒重过的25 ml称量瓶中自然晾干，用万分之一精度的天平进行称重。

1.2.2 气相色谱/质谱联用仪条件

采用美国安捷伦科技有限公司生产的气相色谱/质谱联用仪，型号为6890N型，色谱条件：进样口温度为280 ℃；载气为高纯氦气，流量为1.2 ml/min，线速度为40 cm/s；HP-5弹性石英毛细管柱为30 m×0.25 mm×0.25 mm。质谱仪为5973N四级杆质谱，四级杆温度为150 ℃，离子源为EI源，离子源温度为230 ℃，离子源电离能为70 eV，接口温度为280 ℃，谱库为NIST02L。对于低沸点溶剂低温保存的饱和烃样品，采用的分析条件为：起始温度为40 ℃，恒温3 min，先以3 ℃/min的升温速率升至100 ℃，再以4 ℃/min的升温速率升至290 ℃，恒温30 min。

2 实验结果与分析

2.1 可溶有机质含量

由图1可知，不同层段可溶有机质含量上存在差异：沙四段下亚段样品的可溶有机质含量最低，为0.54~2.34 mg/g，均值为1.20 mg/g；沙四段上亚段样品的可溶有机质含量最多，为4.43~22.64 mg/g，均值为11.38 mg/g；沙三段下亚段样品的可溶有机质含量明显高于沙三段中亚段，为3.92~13.69 mg/g，均值为6.54 mg/g；沙三段中亚段样品的可溶有机质含量较低，为0.25~2.80 mg/g，均值为1.41 mg/g。沙四段上亚段和沙三段中、下亚段样品的可溶有机质含量总体表现出随埋深增加而增加的趋势，而沙四段下亚段样品中可溶有机质含量总体上随埋深变化不明显；沙四段上亚段和沙三段下亚段样品中可溶有机质含量明显高于沙四段下亚段和沙三段中亚段，说明沙四段上亚段和沙三段下亚段的页岩油勘探开发潜力更大，且以沙四段上亚段最好，是最有利的页岩油“甜点”发育区。

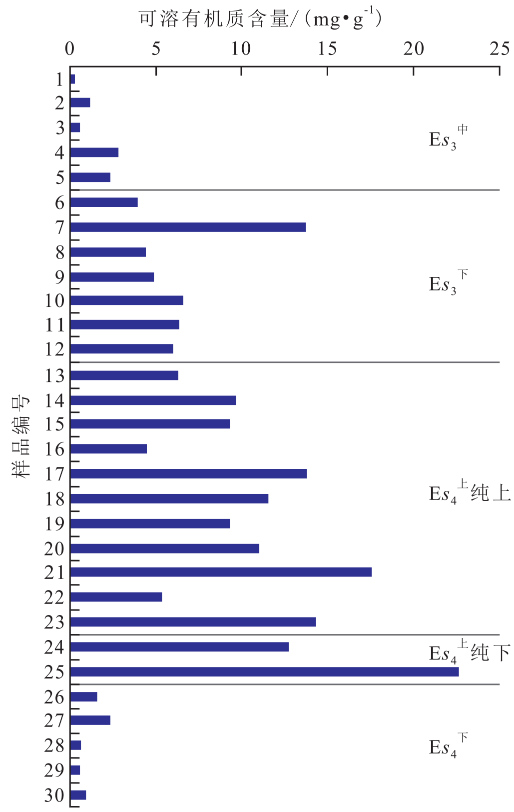

图1 东营凹陷古近系泥页岩样品可溶有机质含量

Fig.1 Total amount of soluble organic matter of Paleogene shale samples from different layers in Dongying Sag

2.2 族组分与流动性

将得到的可溶有机质分离为饱和烃、芳烃、非烃和沥青质4个族组分（图2），各层段不同族组分在含量上存在差异。饱和烃含量呈现出沙四段上亚段最高，含量为2.97~13.46 mg/g，均值为7.53 mg/g；沙三段下亚段次之，含量为2.49~8.49 mg/g，均值为4.19 mg/g；沙三段中亚段与沙四段下亚段相当且较低，沙三段中亚段含量为0.04~1.69 mg/g，均值为0.81 mg/g；沙四段下亚段含量为0.25~1.58 mg/g，均值为0.81 mg/g。芳烃含量呈现出沙四段上亚段最高，含量为0.42~3.80 mg/g，均值为1.78 mg/g；沙三段下亚段次之，含量为0.35~2.09 mg/g，均值为0.89 mg/g；沙三段中亚段较低，含量为0.08~0.41 mg/g，均值

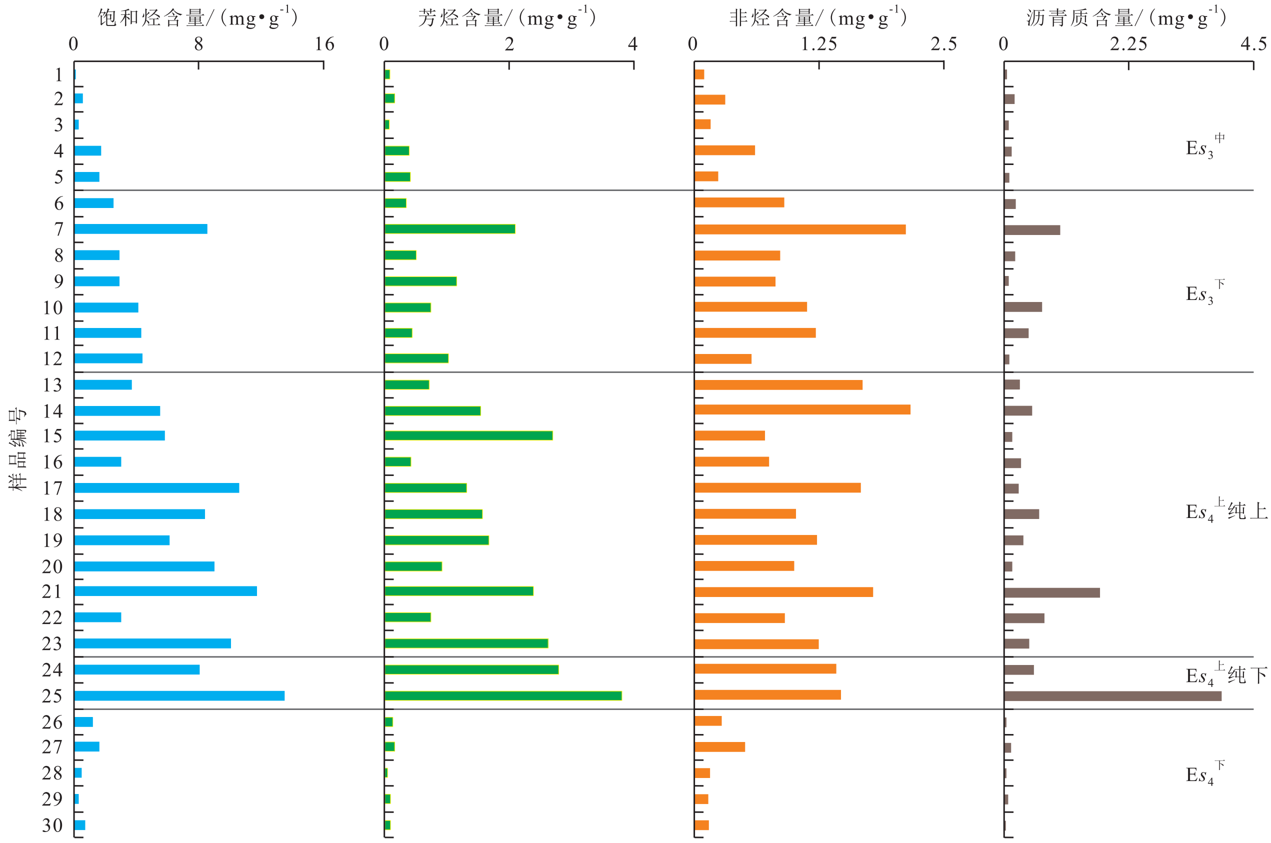

图2 东营凹陷古近系泥页岩样品各族组分含量

Fig.2 Group composition of Paleogene shale samples from different layers in Dongying Sag

为0.22 mg/g;沙四段下亚段最低,含量为0.05~0.15 mg/g,均值为0.10 mg/g。非烃含量呈现出与饱和烃含量一致的趋势,沙四段下亚段含量为0.13~0.50 mg/g,均值为0.24 mg/g,沙四段上亚段含量为0.70~2.16 mg/g,均值为1.31 mg/g,沙三段下亚段含量为0.57~2.11 mg/g,均值为1.08 mg/g,沙三段中亚段含量为0.09~0.60 mg/g,均值为0.28 mg/g。沥青质含量呈现出沙四段上亚段最高,含量为0.13~3.92 mg/g,均值为0.76 mg/g;沙三段下亚段次之,含量为0.07~0.99 mg/g,均值为0.37 mg/g;沙三段中亚段较低,含量为0.04~0.17 mg/g,均值为0.10 mg/g;沙四段下亚段最低,含量为0.02~0.11 mg/g,均值为0.05 mg/g。各层段不同族组分含量基本呈现出与可溶有机质含量一致的趋势,尤其是最为关注的饱和烃,沙四段上亚段和沙三段下亚段样品中饱和烃含量明显高于沙三段中亚段和沙四段下亚段,是页岩油资源更为有利的目标层段,以沙四段上亚段最好,且具有随埋深增加饱和烃含量逐渐增加的趋势,越有利于页岩油的“甜点”发育。

泥页岩经抽提得到的可溶有机质的组分与原油相近,族组分的差异会影响其自身黏度大小,从而影响流动性。不同层段可溶有机质中的族组分占比存在差异(图3),沙四段上、下亚段及沙三段下亚段样品中以饱和烃为主,在可溶有机质含量的占比为46.28%~81.45%(均值为65.21%),芳烃(均值为13.68%)和非烃(均值为15.62%)次之,沥青质占比最低(均值为5.49%),而沙三段中亚段样品饱和烃含量明显低于其他层段(均值为46.74%),最低的1号样品仅占16.92%,且随埋深增加饱和烃含量逐渐增高,非烃含量逐渐降低的趋势。研究认为,原油黏度与其饱和烃含量成反比,与其胶质和沥青质含量成正比[29-36],芳烃有利于沥青质的分散,使胶质与沥青质形成的缔合体能有效地“分散”在饱和烃中,从而有利于降低黏度[31]。此外,带有机官能团且极性强的可溶有机质能够以化学结合的方式牢固的吸附于矿物表面[44-50]。显然,泥页岩中非烃和沥青质是更易化学吸附于矿物表面,使得矿物表面亲油,影响其流动性。因此,泥页岩中饱和烃与芳烃含量的增加有利于页岩油的流动,而非烃和沥青质含量增加会抑制页岩油的流动。由此,可以新定义一个参数——流动性指数(*MI*),其为饱和烃+芳烃与非烃+沥青质的质量比,用以衡量可溶有机质的流动性,*MI*值越大代表可溶有机质流动性越强,反之则越弱。研究区4个层段可溶有机质流动

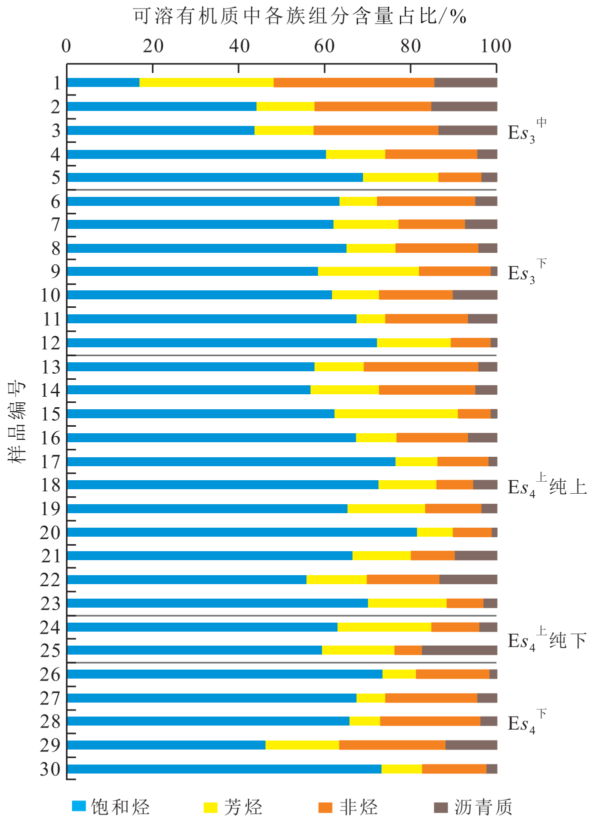

图3　东营凹陷古近系泥页岩样品各族组分含量占比

Fig.3　Percentage composition of group component of Paleogene shale samples from different layers in Dongying Sag

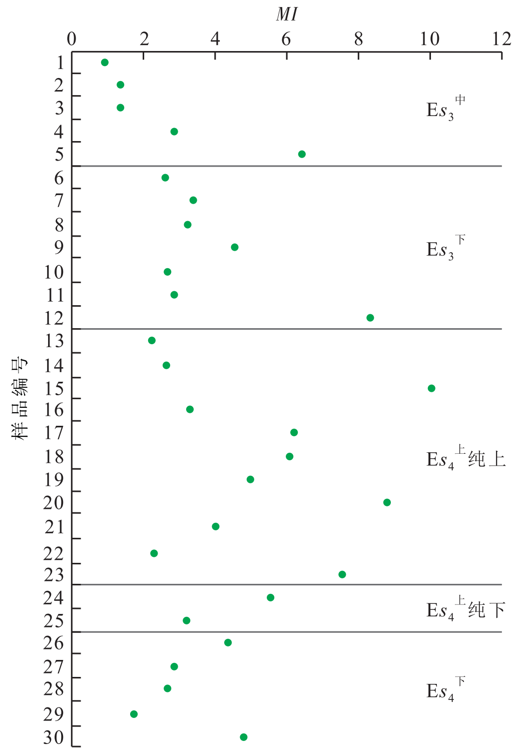

图4　东营凹陷古近系泥页岩样品*MI*分布

Fig.4　*MI* parameter of Paleogene shale samples from different layers in Dongying Sag

性存在差异(图4),*MI*值总体上呈现出沙四段上亚段最高,为2.23~10.20,均值为5.16;沙三段下亚段次之,为2.60~8.32,均值为3.95;沙四段下亚段较低,为1.73~4.81,均值为3.29;沙三段中亚段最低,为0.93~6.41,均值为2.58。因此,沙四段上亚段和沙三段下亚段的页岩油更易流动,尤其是沙四段上亚段。此外,沙三段中、下亚段和沙四段上亚段样品的*MI*值总体表现出随埋深增加而增加的趋势,表明埋藏越深,越有利于页岩油流动。

2.3　饱和烃与流动性

可溶有机质中的液态烃是页岩油开发的重点,可分为轻质烃(C_5—C_{14})和重质烃(C_{15+})[12],而轻质烃由于分子量较低,黏度较小,更易流动、易见产能,是页岩油可有效开采的重要部分[11]。此外,原油中的轻质烃组分可以作为石蜡、沥青质和胶质的溶剂,其含量直接影响蜡、沥青质及胶质的溶解状态,改变蜡晶的形态和分布,对含蜡原油低温流动性的贡献较大[36]。选取各层段代表性样品进行饱和烃色谱(图5)分析,根据饱和烃色谱图可以得到轻质烃和重质烃含量,进而计算出轻质烃和重质烃在可溶有机质中的含量及轻质烃与重质烃比值。本次采用的岩心样品未进行低温封存,尽管在索氏抽提过程中选用正戊烷试剂保护轻质烃,但从饱和烃色谱图中可以看出轻质烃的损失仍较为严重,只能计算出C_{10}—C_{14}轻质烃组分,且损失较多,因此,计算得到的应是样品中残留的轻质烃。从不同层段残留轻质烃含量分布(图6a)来看,总体上呈现出沙三段中亚段最高(均值为12.74%),沙三段下亚段次之(均值为9.20%),沙四段上亚段较低(均值为7.26%),沙四段下亚段最低的趋势(均值为6.82%)。同时,随着埋深增加,残留轻质烃含量呈现出逐渐降低的趋势。重质烃含量总体特征与残留轻质烃不同(图6b),呈现出沙四段下亚段(均值为59.33%)、沙三段下亚段(均值为59.19%)和沙四段上亚段(均值为58.66%)较高且相当,沙三段中亚段最低(均值为39.24%)的趋势。残留轻质烃与重质烃比值总体上的特征与残留轻质烃含量类似(图6c),表现为沙三段中亚段、沙三段下亚段、沙四段上亚段、沙四段下亚段依次降低的趋势,沙四段样品个别有高值。此次得到的是残留的轻质烃含量,无法还原原始的轻质烃含量,但之前对东营凹陷轻质烃含量恢复的研究结果表明[19,25],随埋深和成熟度增加,轻质烃的恢复系数增大,原始含量也呈增加趋势。比较原始轻质烃和残留轻质烃含量(图6d),可以直观的看出轻质烃散失量,对比沙四段上亚段和沙三段下亚段样品发现,沙四段上亚段轻质烃散

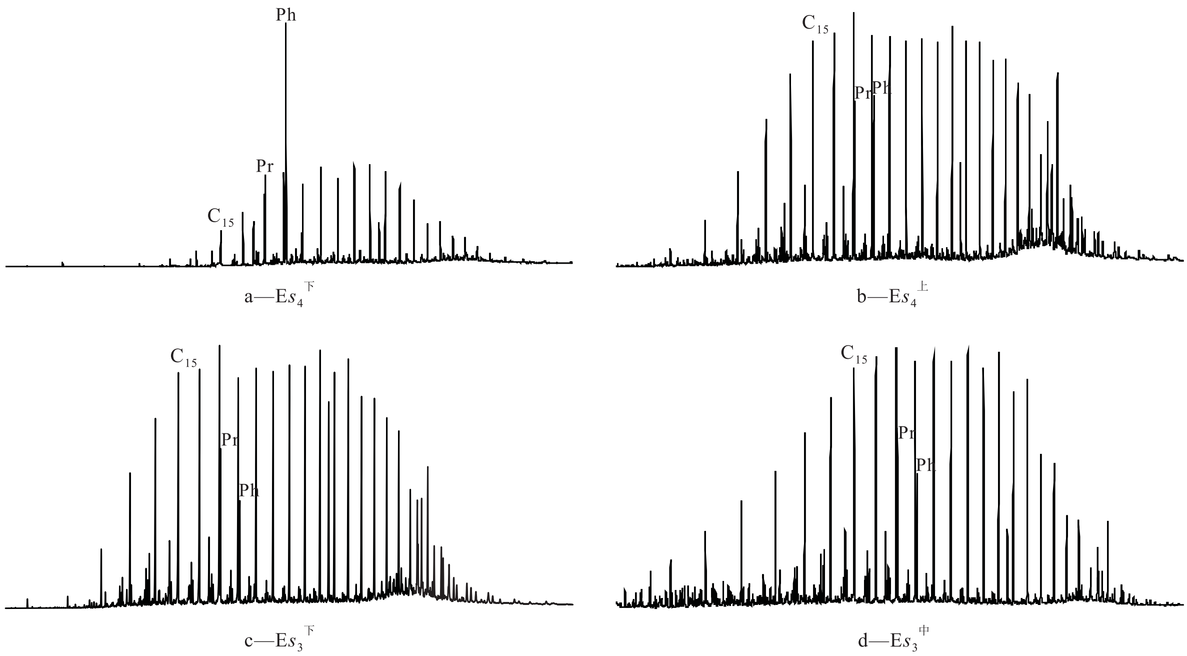

图5　东营凹陷古近系泥页岩代表性样品饱和烃色谱

Fig.5　Saturated hydrocarbon chromatography of Paleogene representative sample from different layers in Dongying Sag

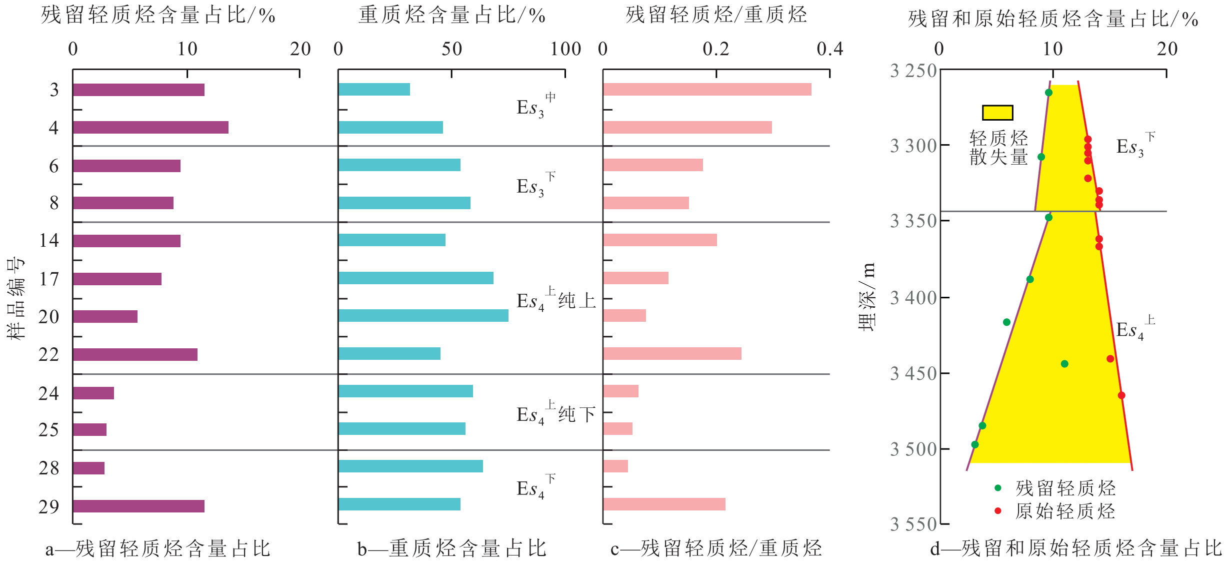

图6　东营凹陷古近系泥页岩样品轻质烃和重质烃含量及轻质烃/重质烃比值(原始轻质烃含量数据来源于文献[23])

Fig.6　Light and heavy hydrocarbon content and light/heavy hydrocarbon ratio of Paleogene shale samples from different layers in Dongying Sag(original light hydrocarbon content data were collected from reference[23])

失量更大。综合对比残留轻质烃、重质烃和原始轻质烃含量特征,发现沙四段上亚段原始轻质烃含量高于沙三段下亚段,且轻质烃散失量更多,这表明从轻质烃角度出发,沙四段上亚段可溶有机质流动性更好,且随埋深增加,流动性越好,越有利于页岩油勘探开发。

2.4　页岩油“甜点”预测

通过对东营凹陷古近系不同层段泥页岩可溶有机质含量、族组成和轻质烃含量等特征的对比,从页岩油物质基础和流动性2方面出发,确定出页岩油“甜点”预测参数,包括反映页岩油丰度的参数(可溶有机质含量及饱和烃含量)及表征页岩油流动性参数(*MI*、轻质烃含量和轻质烃散失量)。首先,在物质基础方面,沙四段上亚段和沙三段下亚段样品中可溶有机质含量及饱和烃含量明显高于沙三段中亚段和沙四段下亚段,以沙四段上亚段更高,同时具有随着埋深增加而逐渐增高的趋势,通过这一参数的对比,可以判定沙四段上亚段和沙三段下亚段是更有利的页岩油发育层段。对比*MI*和轻质烃含量,发现相比于沙三段下亚段,沙四段上

亚段可溶有机质流动性更好。通过以上参数的对比分析，探讨页岩油的物质基础和流动性特征，可以有效预测出页岩油“甜点”区。

3 结论

综合分析东营凹陷古近系泥页岩中可溶有机质特征，确立页岩油“甜点”预测的参数，包括反映页岩油丰度的参数（可溶有机质含量和饱和烃含量）和表征页岩油流动性的参数（*MI*、轻质烃含量和轻质烃散失量）。研究区沙四段上亚段和沙三段下亚段可溶有机质含量和饱和烃含量明显高于沙三段中亚段和沙四段下亚段，是页岩油勘探开发的有利层段，且以沙四段上亚段最好。同时，随着埋深增加，沙四段上亚段和沙三段下亚段可溶有机质含量和饱和烃含量越高，页岩油“甜点”区越发育。相较于沙三段中亚段和沙四段下亚段，沙四段上亚段和沙三段下亚段可溶有机质中*MI*参数较沙四段下亚段和沙三段中亚段更高，同时沙四段上亚段具有更高的原始轻质烃含量和轻质烃散失量，流动性更好。同时，沙四段上亚段和沙三段下亚段*MI*和原始轻质烃含量总体呈现出随埋深增加而增加的趋势，表明埋藏越深，越有利于页岩油流动。综合分析认为，东营凹陷沙四段上亚段和沙三段下亚段是页岩油有利的发育层段，以沙四段上亚段更好。

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