

碳同位素录井技术发展现状及展望

牛 强^① 瞿煜扬^{②③} 慈兴华^① 周文治^④ 张焕旭^{③⑤}

(^①中石化胜利石油工程有限公司地质录井公司;^②苏州冠德能源科技有限公司;

^③苏州加州能源与环境研究院有限公司;^④长江大学石油工程学院;^⑤北京大学工学院)

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摘 要 在传统的油气勘探开发中,碳同位素能够确定油气成熟储集层与烃源岩的关系、区分油气混合来源由于页岩气成分简单,组分中的同位素信息尤为重要。随着同位素分析技术快速发展,国内外开始尝试在现场发展实时同位素录井技术,各大油服、仪器公司相继研发出同位素录井设备,在现场进行测试。碳同位素录井是一种在油气勘探发现现场的快速、连续、准确、经济获取同位素信息的新型录井技术,对地质条件下油气藏的发现和检测提供了实时和连续的地球化学表征,为录井工作提供了全新的数据来源和数据解释。碳同位素录井技术已逐渐被国内油气勘探开发单位所接受,以东营和南川两口井的碳同位素录井为例,总结碳同位素录井技术的起源及发展、碳同位素录井方法及其优势,并展望该技术未来更加广阔的应用前景。

关键词 页岩气 碳同位素 碳同位素录井 色谱—光谱联用仪 甜点识别

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0 引 言

随着经济发展,对天然气的需求缺口迅速扩大并投入大量资金用于页岩气的开发,这也促进了油气勘探开发技术与装备的升级,录井行业将发挥越来越重要的作用。碳同位素录井作为一种新型录井技术,近年来被引进国内,在环渤海地区和四川盆地的油气田多口钻井尝试碳同位素录井,均取得了不错的效果。本文通过总结碳同位素录井技术的发展历史和技术原理,结合实例介绍碳同位素录井技术在油气勘探开发中的应用。

1 谱仪技术的发展

1.1 同位素质谱技术和质谱小型化的发展

1919年英国的汤姆孙和阿斯顿研发成功第一台质谱仪^[1],通过离子源将待测物质分子离子化,经过电场加速进入质量分析器,带电粒子在扇形磁场中发生偏转,并通过电场使具有不同质荷比的离子分开进入离子检测器,实现了同位素的测量。质谱技术很快成为一种常规的分析手段,被化学家广泛

采用。常见的质谱仪按质量分析器可分为扇形磁场质谱仪、四极杆质谱仪、飞行时间质谱仪、离子阱质谱仪等。离子源按电离方式主要有电轰击(EI)、化学电离(CI)、激光电离(MALDI)和电喷雾(ESI)等^[2]。离子检测器主要为电子信号放大器。

当前质谱分析技术已十分成熟,在不同领域得到广泛的应用,是测量同位素的最常用方法^[3],其主要的发展方向有两个,一是超高精度测量,二是质谱仪器的小型化。如SPEAR公司推出了应用于野外的Scanning Q-MS同位素测量仪器,但其性能与实验室常用的同位素质谱仪相比还有较大差距。由于质量分析器的原理对压力即高真空度的苛刻要求^[4-5],以现有的元器件水平,质谱仪整体结构的进一步优化受到很大的限制。

1.2 同位素光谱技术的发展

1814年,德国光学家夫琅和费制成了第一台分光镜^[6],1859年基尔霍夫和本生发现物质的光谱线在光谱中同时呈现,彼此并不互相影响^[7],据此可通过元素的光谱特征判别化学元素。后来将衍射光栅作为分光元件制成光谱分析仪,使分辨率大为提高,

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牛强,高级工程师,1975年生,中国石油大学(北京)博士毕业,现在中石化胜利石油工程有限公司地质录井公司从事综合地质和录井技术推广应用等研究工作。通信地址:257064 山东省东营市东营区乐园路1号。电话:(0546)8621357。E-mail:slniuq@126.com

现代光谱分析技术开始快速发展。

1984年Andreson等首次将光腔衰荡光谱技术应用于测量低损耗高反射膜的反射率^[8],以基于测量衰减率而不是绝对吸收的原理使其较传统吸收光谱拥有更高灵敏度,PICARO公司进一步将其发展为波长扫描光腔衰荡光谱(WS-CRDS)技术,实现较高精度的稳定同位素测量,同时美国LGR公司将积分腔输出光谱技术(ICOS)改进为偏轴积分腔输出光谱技术(OA-ICOS),在具有同等水平的灵敏度情况下,则光腔衰荡光谱更适合用于实地测量^[9]。

近年来,可调谐半导体吸收光谱(TDLAS)和中红外激光吸收光谱技术的应用逐渐成熟^[10-12],实现了稳定同位素痕量样品的高精度测量,TDLAS的代表性仪器为美国CAMPBELL公司的TDL-TGA 100。基于中红外激光吸收光谱技术的代表性仪器有美国THERMO公司的Delta Ray CO₂比值同位素红外光谱,其光源为中红外激光差频发生器(DFC);美国Peeri公司的AGT 2000碳同位素测量仪,其光源为量子级联激光器(QCL)。由于中红外光吸收光路短,绝大部分气体分子均对中红外波段有较好吸收的特性^[13],上述两种类型仪器均实现了对痕量样品连续或非连续的高精度测量,精度达到质谱仪的同等水平。

此外还有20世纪90年代以来快速发展的傅里叶变换(FTIR)红外光谱技术,这种不同于红外吸收光谱的干涉型光谱技术,具有高光通量、低噪音、测量速度快、分辨率高、波数准确度高、光谱范围宽等优点。

总体来说,光谱仪较质谱仪更为适应样品类型、浓度大幅度变化以及工作环境不够稳定的实地测量,易于维护,也更加经济,有着更为广阔和多样化的应用前景。

2 同位素录井技术的起源与发展

在传统的油气勘探开发中,碳同位素在地质事件分析、油源对比、成熟度研究、沉积环境分析和古环境分析中均有较好的应用^[14-15],目前的质谱单体同位素测量仪器GC-IRMS价格昂贵,维护复杂,对环境稳定性要求很高。因此,同位素分析还多停留于现场取样实验室分析的工作模式,这极大地限制了其在油气勘探中的应用。

20世纪90年代国外油气工作者尝试在钻井现场批量采集钻井液气,送回实验室分析,针对研究天

然气成因^[16]、判断储集层封闭性^[17]及识别断层构造等做了许多工作。1999年Ellis提出“Mud gas isotope logging while drilling”的概念^[18],并从2002年开始首次将碳同位素录井(MGIL)大规模运用于区域油气成藏和生产评价中,其在近三年的时间里在18口钻井中开展了同位素录井工作,在没有电测和MDT数据的情况下,利用MGIL数据发现和识别了砂岩体中的断层和隔层^[19],对于认识储集层物性和后续的开采具有重要意义,美国油气界由此开始重视碳同位素录井工作。2005年斯伦贝谢旗下的GEOSERVICES(法国地质服务公司)、英国的CSS公司、加拿大ISOMETRIC公司及美国的LGR公司先后推出了基于光谱原理的井口碳同位素实时录井仪,实现了C₁同位素的现场实时测量,让同位素技术真正进入了现场,但由于吸收池体积大而不能和色谱联用,导致无法分析钻井液气中的C₂、C₃,也无法测量浓度较低的罐顶气样品,这在很大程度上限制了井口同位素录井的功能。

2013年SPEAR公司对传统质谱进行了简化和改进,研发出了基于色谱联用和扫描质谱原理(Scanning Q-MS)的同位素录井仪,实现了C₁、C₂、C₃的测量,精度达到±1%,但由于质谱质量选择器仍存在对高真空、低干扰环境的要求,无法达到更高的精度。GEOLOG公司和ISOTECH公司基于Picarro CRDS开发了相应的光谱同位素录井仪,同时PEERI旗下的AGT公司开发空心波导和量子级联激光器技术(HWG+QCL)^[20-21],推出了色谱—质谱联用同位素录井仪(GC-IR²),解决了传统光谱光腔大、检测范围窄的问题,同位素录井技术开始在北美页岩气产区广泛应用。

法国地质服务公司最早将碳同位素录井技术引进中国,在南海西部琼东南盆地乐东凹陷YC-1-1-1井开展了甲烷同位素录井,根据甲烷同位素值判别烃源岩的成熟度和天然气类型^[22],国内录井行业开始认识到碳同位素录井的重要价值。

常见光谱碳同位素录井仪的工作原理为:将脱气器析出的气体经过处理装置后与同位素录井仪器串联,通过快速色谱将烃类气体按组分进行分离,依次进入氧化池燃烧成为CO₂,再进入中红外激光光谱测量腔内,利用¹²C-O、¹³C-O分子键对激光的吸收特征峰不同,实现碳同位素的测量。

3 碳同位素录井工作流程

针对页岩气同位素现场实时检测,碳同位素录

井的样品主要为 Isopack(钻井液气)和 Isojar(岩屑罐顶气)。钻井液气是钻井液循环带上的地层破碎释放气,反映了钻遇地层最为直观的同位素信息,通过取样管在脱气器处采集保存,手动注样测量,或将碳同位素录井仪与脱气器连接,经过处理装置后实现自动进样品;岩屑罐顶气实际上是岩屑解析出来的气体,经钻井液池振动筛处捞拾岩屑,洗净装罐、加水密封后倒置,气体通过纳米孔隙逸散产生同位素分馏效应,通过同位素分馏的程度与速率可以反映页岩含气量与纳米孔隙网络发育特征(图1)。由此获得连续、立体的碳同位素信息,结合区域成藏背景等相关地质信息,进行油气成藏组合特征和油气富集特征的识别。

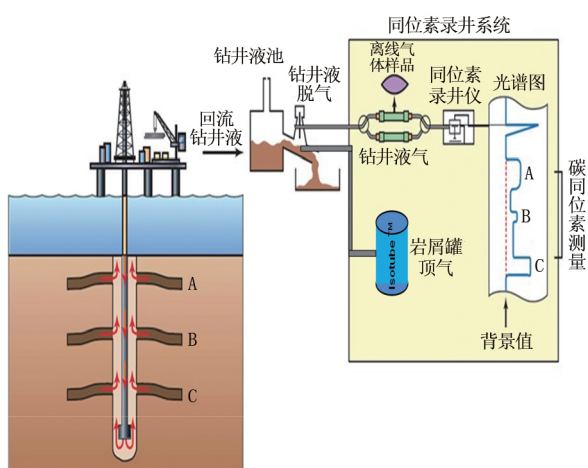


图1 碳同位素录井工作流程

4 碳同位素录井技术应用实例

4.1 东营凹陷某井

民丰洼陷位于东营凹陷的东北部,其北部为陈家庄凸起,南邻中央隆起带,东部为青坨子凸起,西部为利津洼陷。该井位于民丰洼陷北部斜坡,该区域的沙四下亚段发育多期近岸水下扇,扇体紧邻生油中心分布,与湖相烃源岩呈指状接触,是良好的油气聚集场所,但因近岸水下扇砂砾岩体混杂堆积,沉积厚度大,非均质强,物性差,测井响应不明显,同时由于埋藏深,上覆巨厚的盐膏层,盐下地震资料品质变差,砂砾岩扇体不能进行精确刻画^[23-25]。

图2展示了该井垂深方向碳同位素变化趋势。井段1900~3300 m 碳同位素值逐渐变重,且 C_2 、 C_3 同位素值变化趋势平缓,与该地层烃源岩干酪根处于成熟或成熟早期阶段的特征相匹配,同位素偏轻与母质的有机质类型有关;在 3500 m 以下 C_1 同位

素值明显变重,脱离了正常的同位素随深度变化序列,结合相关层位和构造信息,推测这种分馏是深部高成熟原油裂解气顺砂体向上运移的结果,同时 3500 m 以下的沙四下亚段气油比明显高于上部也印证了这种推测。由此进一步推测该区域 3500~4000 m 的沙四下亚段可能存在一个地质甜点。

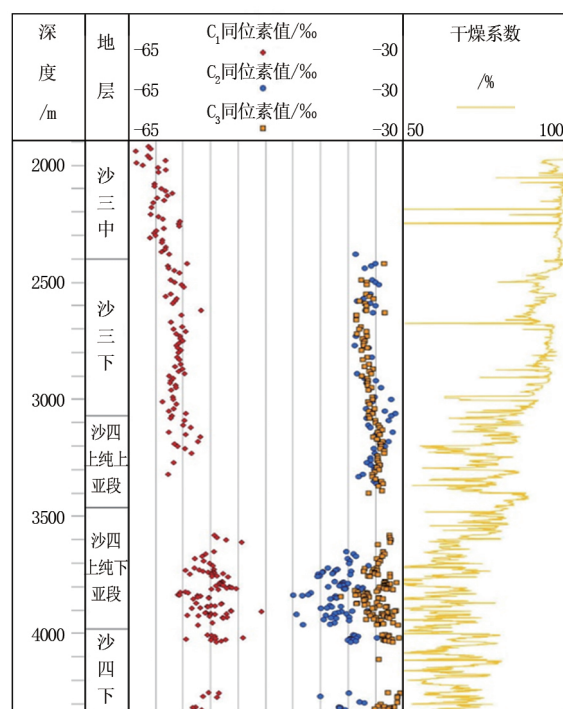


图2 东营某井沙三、沙四段钻井液气碳同位素剖面

4.2 四川某海相页岩气井

该井位于四川盆地的东南缘,下古生界的五峰—龙马溪组黑色页岩在区域内广泛发育,厚度稳定,是区域内的一套重要烃源岩,也是四川盆地页岩气勘探的重点层位^[26]。研究表明,该地区干酪根类型主要为Ⅱ1型和Ⅱ2型,有机质成熟度高,微观孔隙发育,与北美页岩相似^[27],为页岩气勘探的有利区域,但页岩具有“自生自储”的特点^[28],孔隙过于微小、孔隙连通性、岩性差异小导致油气释放缓慢,往往造成录井过程气测值低,容易错过重要产油层。考虑到页岩气“自生自储”的特点,烃类的碳同位素对母质有较好的“继承性”,烃类的“扩散”或“混合”会留下特定的“同位素痕迹”,开展了同位素录井工作以分析该区域的页岩气富集特征与地质甜点属性。

针对钻井液气进行碳同位素录井,核心层段 2 m/样,非核心层段 5 m/样,涵盖了小河坝组、龙马溪组、五峰组,得到完整的同位素录井曲线(图3)。 C_1 的同位素值整体在 $-33‰ \sim -30‰$ 之间, C_2 的

同位素值整体在 $-35\text{‰} \sim -31\text{‰}$ 之间,出现了同位素倒转的特征,这跟我国南方海相五峰—龙马溪组页岩气及北美某些页岩区普遍出现同位素倒转的情况相符^[29-30]。相关统计表明同位素倒转与页岩气的高产往往具有较好的对应关系^[31],通常认为原油裂解气的混入造成了同位素倒转^[32-33]。该井龙马溪组页岩同位素与焦石、彭水、丁山区块同位素值较为相近,说明其与盆地东南区块的页岩有较为相似的母质类型和向上排烃类过程,龙三段和小河坝组气测值较低,湿度偏高,此处的烃类可能是下部排出的液态烃裂解的产物。在龙一段下部 C_1 和 C_2 的同位素值分别为 $-31\text{‰} \sim -30\text{‰}$ 、 $-35\text{‰} \sim -33\text{‰}$,同位素倒转程度趋大。

针对岩屑进行罐顶气碳同位素录井,核心层段

5 m/样,非核心层段 20 m/样,得到钻井液气和岩屑罐顶气的同位素值对比(图 4),发现岩屑罐顶气同位素值在 2750 m 以下分馏明显。北美页岩气产区大量的数据统计显示,同位素分馏明显的区段往往是页岩气的“甜点”段^[31],碳同位素分馏特征明显指示了页岩气储集层高压、高含气量和良好的孔隙结构特性。

结合地质背景和成藏背景分析,给出如下结论:龙三段上部和小河坝组上部的气态烃是下部烃源岩排烃过程液态烃裂解的产物,可以结合相关地层岩性、裂缝、构造油藏的条件,寻找可能存在的次生气藏;在 2750 m 以下钻井液气同位素倒转程度加大、岩屑罐顶气的同位素分馏明显,推测存在页岩气“甜点段”,应作为重点勘探层位。

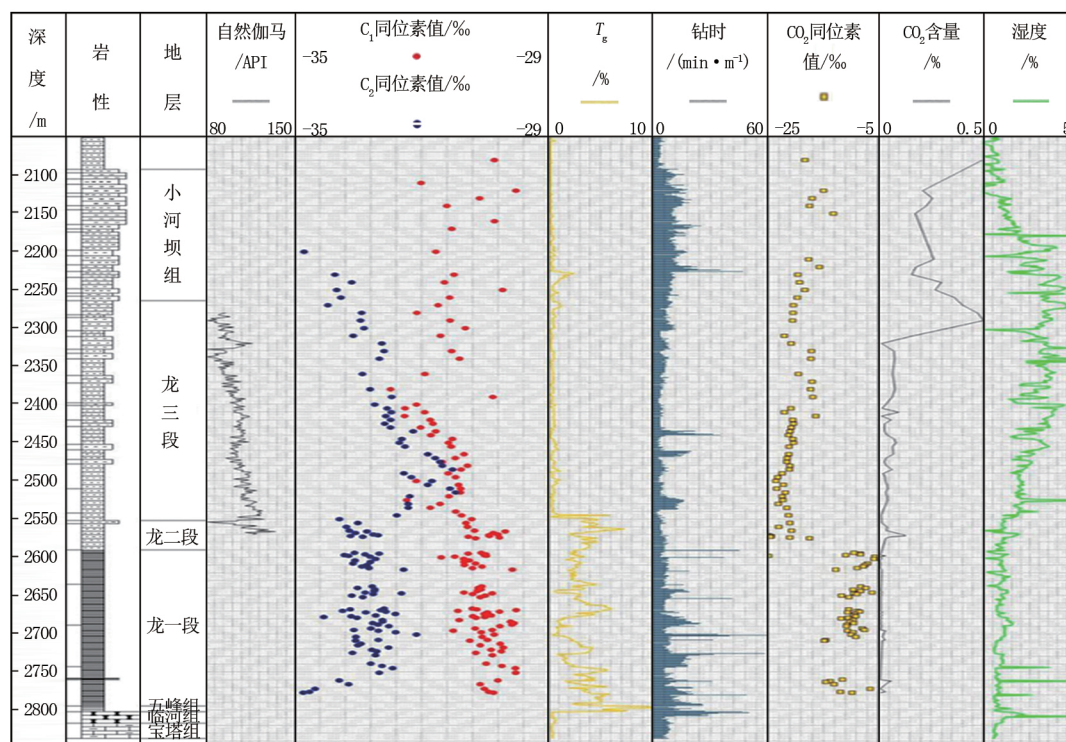


图3 四川某页岩气井碳同位素录井成果

4.3 碳同位素录井技术应用总结

碳同位素录井仪器本身就是一个高精度的 C_1 — C_8 轻烃色谱分析仪,与钻井过程中钻遇烃类碳同位素值的实时性检测结合,可提供有关烃类来源、特征和相态的重要信息。稳定碳同位素值与油气的生成和来源存在直接关系,涵盖了烃源岩中有机质来源、有机质类型、有机质成熟度等重要地球化学信息,可直接判定油气的烃源层,并为油气成藏组合的分析和产油层的判别提供直接证据。

碳同位素在反映油藏连通性和混合作用中也具优势,能给评价油气藏的盖层有效性和断层封闭性提供有效的解释手段,从而完善了地质流体分析解释的方法。针对页岩气产区,碳同位素的单调性变化也能作为GR、R等波形导向的补充来识别隐形断层。由于页岩气的分馏速率与地层压力、含气量、孔隙度、渗透率存在较强的相关性,通过测量同位素值的分馏,可进行气田产气量的预测和给出分段压裂方案的建议。

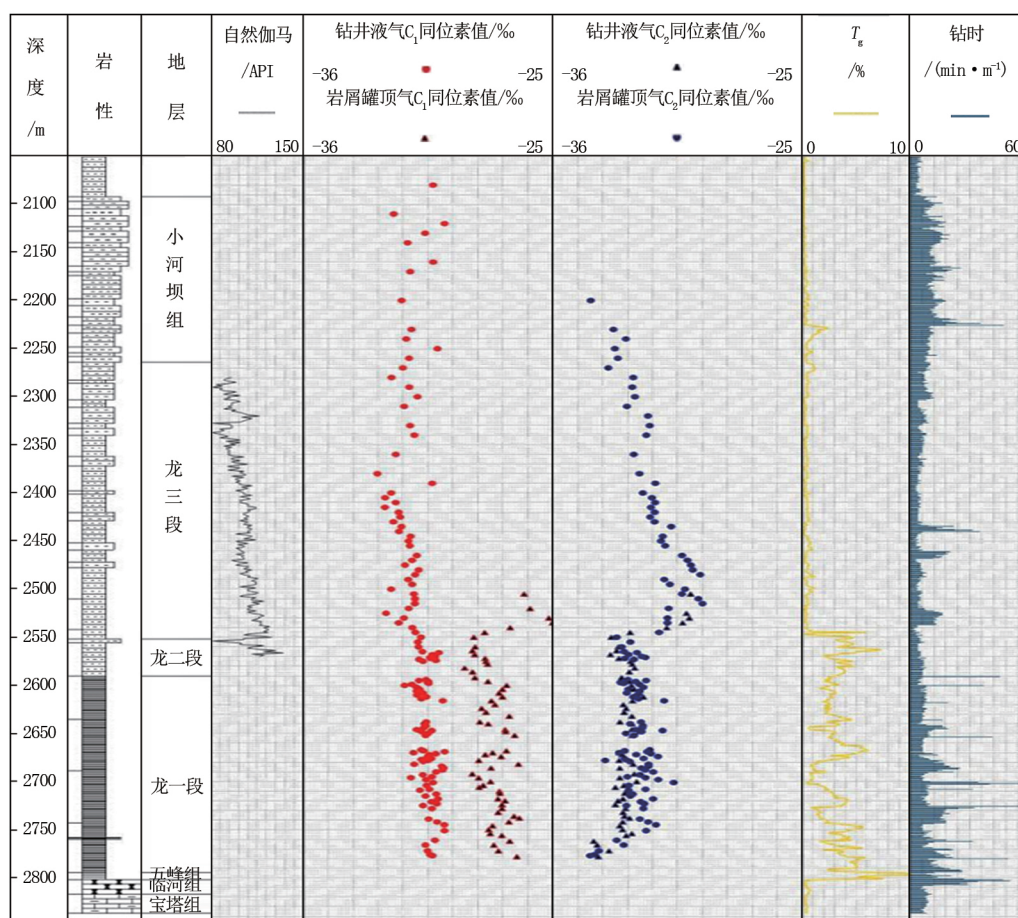


图4 岩屑罐顶气和钻井液气同位素剖面

5 碳同位素录井技术的展望

2017年中国页岩气年产量突破了 $90 \times 10^8 \text{ m}^3$ ，“十三五”规划提出了2020年实现页岩气年产量达到 $300 \times 10^8 \text{ m}^3$ 的目标，国内对页岩气的需求旺盛且在国家大力支持下，页岩气勘探进入战略机遇期，从而对勘探开发提出了更高的要求，油服公司纷纷加大研发力度，这也促进了碳同位素录井设备的升级、革新。碳同位素录井技术的发展方向有三个。

(1) 实现超高精度的同位素测量。最新中红外吸收光谱技术的测量精度在原理上已经能达到传统质谱仪 GC-MS 的水平，但受限于现场条件，如电压、温度等条件，只能接近质谱仪的测量精度，略有差距，需要进一步提高元器件性能，优化整体结构，提高抗干扰能力，如采用双通道、三通道检测器提高检测精度，为现场测试提供实验室级别的数据。

(2) 拓宽测量范围，不局限于碳同位素。未来可以考虑将 H、N、O 同位素的测量也纳入到同位素录

井体系中。天然气中 CO_2 碳同位素值能用来判别是无机成因还是有机成因^[34-36]，而 N_2 、 H_2 的同位素值变化较大，成因复杂，其对天然气成因和母质来源有一定的指示作用^[37-45]，但利用 N、H 同位素值进行成因分析还需要进一步的研究，现阶段往往需要其他指标辅助判断，油气勘探开发中对这三种气体的研究尚不够重视。由于红外吸收光谱在原理上可以测量这三种气体的 C、N、H 同位素值，多种元素的同位素数据相互印证，将为油源对比、成熟度研究、沉积环境分析带来更多的数据来源，现场的同位素测量将真正起到“DNA 识别”的作用。

(3) 同位素录井设备和其他仪器的集成化。在井场与热解仪、气测录井仪等配套使用，将大幅提高碳同位素数据的使用效率和价值。国内某碳同位素录井设备厂家将同位素录井仪与前处理装置及相关仪器集成到一辆录井车中，可处理和测试的样品类型更加多样化，车载系统保证了较为稳定的工作环境，且具有高机动性，移动同位素录井房也是移动的

同位素实验室,可快速部署到井场开展工作。

6 结 论

碳同位素录井是一种在油气勘探发现现场快速、连续、准确、经济获取同位素信息的新型录井技术,对地质条件下油气藏的发现和检测提供了实时和连续的地球化学表征,为录井作业提供了全新的数据来源和数据解释,在判断油气藏成因、类型,反映油气藏连通性和混合作用中具有优势,也能给钻井和地质导向、压裂层段的选取、油气勘探综合分析及未来井位设计等提供重要的数据参考。碳同位素录井技术引入后,珠江口盆地、琼东南盆地、四川盆地页岩气产区均开展了碳同位素录井并积累了大量经验,未来在包括四川盆地在内的页岩气产区,碳同位素录井有着更加广阔的应用前景。

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Application of evaluation technique of bit efficiency while drilling. Hu Zongmin, Yuan Boyan, Zhang Xiangguo, Han Bingbing, Li Yi, Lai Fubin and Li Yun. *Mud Logging Engineering*, 2019, 30(3):1-7

In the bit efficiency evaluation using R • Teale mechanical specific energy model and bit feed rate, although the bit weight and torque in the mechanical specific energy model are corrected, there are still some problems in data processing, which makes it very difficult to use the traditional crossplot chart for drilling engineering guidance. Therefore, the intersection curve of mechanical specific energy and bit feed rate is drawn through the steps of data interpolation, data screening, deletion and smoothing filtering to the corrected mechanical specific energy data. By combining the shape and area of the intersection curve with the drilling condition, the rock breaking efficiency and bit behavior can be judged, and engineering analysis of bit balling, drill string vibration, bit passivation, bit lectotype and so on can be carried out. The evaluation examples of bit efficiency while drilling in drilling site prove that this method has better real-time performance and effectiveness, which can reduce drilling engineering accidents, shorten drilling cycle and reduce drilling cost.

Key words: mechanical specific energy, evaluation while drilling, feed rate, curve intersection, bit efficiency

Hu Zongmin, No.1 Geo-Logging Company, Daqing Drilling & Exploration Engineering Corporation, Daqing City, Heilongjiang Province, 163411, China

Development status and prospect of carbon isotope logging technology. Niu Qiang, Qu Yuyang, Ci Xinghua, Zhou Wenzhi and Zhang Huanxu. *Mud Logging Engineering*, 2019, 30(3):8-15

In traditional oil and gas exploration and development, carbon isotopes can determine the relationship between mature oil and gas reservoirs and source rocks and distinguish the mixed sources of oil and gas. Due to the simple composition of shale gas, isotopic information in the composition is particularly important. With the rapid development of isotopic analysis technology, domestic and foreign began to try to develop real-time isotopic logging technology in the field. Major oilfield service and instrument companies have developed isotope logging equipment and conducted the test in the field. Carbon isotope logging is a new mud logging technology which is rapid, continuous, accurate and economical to obtain isotopic information in the field of oil and gas exploration and development. It provides real-time and continuous geochemical characterization for the discovery and detection of hydrocarbon reservoirs under geologic conditions, and provides a new data source and data interpretation for mud logging work. The carbon isotope logging technology has been gradually accepted by the domestic oil and gas exploration and development units. Taking carbon isotope logging of two wells in Dongying sag and southern Sichuan Basin as examples, the authors summarized the origin and development of carbon isotope logging technology, carbon isotope logging method and its advantages, and looks forward to its broader application prospects in the future.

Key words: shale gas, carbon isotope, carbon isotope logging, chromatography-spectrum coupling instrument, high-quality reservoir identification

Niu Qiang, 1 Leyuan Road, Dongying District, Dongying City,

Shandong Province, 257064, China

Identification and interpretation of formation water by gas logging data. Cui Jian, Wang Lei, Li Jiaqi, Jin Qiuying and An Yi. *Mud Logging Engineering*, 2019, 30(3):16-20

Oil, gas and water rarely exist alone in the reservoir, and most of them exist in the form of two-phase or three-phase coexistence, so it is necessary to accurately analyze the water signature of the formation and eliminate the interference of formation water to gas logging interpretation. The fault blocks in Jidong Oilfield are broken, there are many types of traps, and the oil, gas and water distribution are complex, especially in Gaoshangpu, Liuzan and Bogezhuang blocks which have been waterflooded, the distribution of subsurface fluid is more complex and the identification is more difficult. In the process of gas logging interpretation, the emphasis is laid on analyzing whether the formation contains water or not, which solves the problem of unclear identification of formation water cut in gas logging interpretation. Practical application in 23 layers of 10 wells in Jidong Oilfield shows that the interpretation coincidence rate reaches 86.96%, which effectively improves the interpretation level of field gas logging.

Key words: formation water, gas logging, gas parameter, fluid property

Cui Jian, Engineering Supervision Center of Jidong Oilfield, Tanghai County, Tangshan City, Hebei Province, 006320, China

Analysis of influencing factors and control measures of three-dimensional quantitative fluorescence logging. Jiao Xiangting, Sun Fenglan, Yang Jianhua, Zhao Huixia, Wang Yufang, Yue Ying and Lan Xingmin. *Mud Logging Engineering*, 2019, 30(3):21-26

Three-dimensional quantitative fluorescence logging technology has been widely used in oil exploration and development, but when applied to shale oil samples, it is found that there is a big error in analytical data when it is operated by conventional methods. In order to improve the quality of three-dimensional quantitative fluorescence logging data, the factors that affect the accuracy of the analytical data and spectrogram were found out by analyzing the original data. Furthermore, experimental analysis was made to the factors such as impurity in shale oil sample, soak time of sample, diluted concentration of sample and change of instrument light source, etc. By adopting a unified standard substance, the calibration method of three-dimensional quantitative fluorescence instrument was established, the rational soak time was determined, and the software was modified and perfected to ensure that the diluted concentration of the sample is kept in a reasonable range. After taking the above control measures, the error rate of three-dimensional quantitative fluorescence analysis data and spectrogram decreased from 5.02% to 1.23% in the real well application process, and the effect is remarkable.

Key words: three-dimensional quantitative fluorescence, analysis principle, influencing factor, soak, dilute, light source, control measure

Jiao Xiangting, No.1 Mud Logging Company, BHDC, Tuanjie East Road, Dagang Oilfield, Tianjin, 300280, China