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移动扫码阅读

新疆高钠煤中钠元素的地球化学研究现状

杨承伟[1,2],丁　华[1,2],白向飞[1,2],唐跃刚[3],何　金[1,2],袁东营[1,2]

(1.煤炭科学技术研究院有限公司 煤化工分院, 北京　100013; 2.煤炭资源高效开采与洁净利用国家重点实验室, 北京　100013;3.中国矿业大学(北京) 地球科学与测绘工程学院, 北京　100083)

摘　要:新疆煤炭资源丰富,以低灰、低硫、低磷的发电和化工用煤为主,已成为我国煤炭开发利用的主战场。但由于特殊的地质环境和地下水作用,新疆部分煤层中富集了大量钠元素,引起的锅炉结渣结垢问题一直制约着高钠煤的清洁高效利用。新疆高钠煤及其中钠的分布赋存特征成为近 10 年来煤化学基础研究领域的一个热点。系统总结了煤中钠赋存状态研究成果,概述了煤中钠含量的测定方法,分析了控制煤中钠富集的关键地质因素,并讨论了煤炭利用过程中钠的迁移规律及其影响因素。研究表明:煤中钠的赋存状态以无机钠为主,少量为有机钠,逐级化学提取结果进一步表明煤中的钠以水溶态为主。目前对煤中钠含量的测定主要是通过燃烧和一定条件下的灰化、消解后进行的,但针对于新疆煤中钠以水溶态为主要赋存状态这一基本事实,逐级提取法可以减少样品前处理中钠的损失,进而更准确地反映煤中钠的含量。煤中钠的富集成因主要包括生物成分累积、碎屑沉积物输入、泥炭孔隙中溶液沉淀,以及成煤后期海水作用等。以往国内外对高钠煤的研究比较关注海水侵入的影响,但新疆的高钠煤中钠的富集需要重点考虑地表水淋滤和地下水驱动因素。在煤炭加工利用过程中,钠的赋存形式和煤炭利用方式决定了其释放途径。深入研究煤中钠元素富集的地球化学控制机理以及煤炭燃烧等过程中钠的迁移机制,对高钠煤的合理开发利用意义重大。

关键词:高钠煤;钠元素;地球化学;赋存特征;成因与控制;迁移与释放

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Geochemical research status of sodium element in high-sodium coal in Xinjiang

YANG Chengwei[1,2],DING Hua[1,2],BAI Xiangfei[1,2],TANG Yuegang[3],HE Jin[1,2],YUAN Dongying[1,2]

(1.*Coal Coal Chemical Industry Branch of CCTEG China Cool Rersearoh Institute*,*Beijing*　100013,*China*; 2.*State Key Laboratory of Coal Mining and Clean Utilization*,*Beijing*　100013,*China*; 3.*College of Geoscience and Surveying Engineering*, *China University of Mining and Technology Beijing*, *Beijing*　100083, *China*)

Abstract:The coal resource in Xinjiang Uygur Autonomous Region, China, which is dominated by low-ash, low-sulfur, and low-phosphorus coals for power generation and chemical industry, is abundant and has become the main battlefield for coal development and utilization in my country. However, due to the special geological environment and the action of groundwater, a large amount of sodium is enriched in some coal seams in Xinjiang. The clean and efficient utilization of the high-sodium coal in Xinjiang has been constrained because of slagging and fouling. Xinjiang high-sodium coal and its distribution and occurrence characteristics of sodium have become a hot spot in the field of basic coal chemistry research in the past decade. This paper systematically summarized research results of the occurrence state of sodium in coal, summarized the determination method of sodium content in coal, analyzed the key geological factors controlling sodium enrichment in coal, and discussed the migration law of sodium and its influencing factors in the process of coal utilization. Previous research demonstrated that sodium in coal dominantly occurs as inorganic forms, with minor organic forms. The sequential chemical extraction further indicated that sodium in coal is mainly water-soluble. At present, the determination procedure for coal sodium is conducted

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作者简介:杨承伟(1993—),男,青海西宁人,助理研究员,博士。E-mail:ycweduyong@163.com
通讯作者:丁　华(1980—),女,黑龙江鸡西人,副研究员。E-mail:briccding@126.com

through combustion and ashing and digestion under certain conditions. However, because sodium in Xinjiang coal is mainly water-soluble, a sequential extraction can reduce the loss of sodium in sample preparation and the sodium content can be detected more precisely.The enrichment of sodium in coal mainly includes the accumulation of biological components, the input of clastic sediments, the precipitation of solution in peat pores, and the action of seawater in the later stage of coal formation. Previous studies on high-sodium coal at home and abroad focused on the influence of seawater intrusion,but the enrichment of sodium in high-sodium coal in Xinjiang needs to focus on the surface water leaching and groundwater driving effect. In the process of coal processing and utilization, the occurrence form of sodium and the way of coal utilization determines its release pathway. In-depth study of the geochemical control mechanism of sodium enrichment in coal and the migration mechanism of sodium in the process of coal combustion is of great significance for the rational development and utilization of high-sodium coal.

Key words: high-sodium coal; sodium element; geochemistry; occurrence characteristics; genesis and control; migration and release

0　引　言

煤的组成、结构与性质对煤炭清洁高效绿色的发展有重要影响。由于煤炭储量丰富,煤质优良,新疆已成为我国煤炭清洁高效利用的主战场之一,其中准东矿区属于我国超大型煤产区[1-2],目前已经探明的煤炭储量超过 2 000 亿 t,预测储煤量达 3 900亿 t[3]。新疆煤具有低灰、低硫、低磷等优点[4-7],是良好的发电和煤化工用煤。但由于特殊的地质环境和地下水作用,煤层中富集了大量钠元素,煤灰中的氧化钠含量达 3%~8%,部分区域超过 10%,在很大程度上影响了其利用性能。我国目前所报道的高钠煤均产自新疆,尤其是准东矿区产量巨甚。国际上也有过对高钠煤的报道和研究:土耳其 Cayirhan 煤田的煤层因受到火山碎屑物质的输入,导致煤中高度富集含钠、钙的沸石[8];乌克兰西顿巴斯石炭纪高钠煤、德国鲁尔 Frotuna-Garsdorf 褐煤和澳大利亚高钠褐煤,均因沉积环境偏海相导致钠含量较高[9-14]。目前国内外对“高钠煤”尚无统一定义,按照现行煤炭行业标准 MT/T 1074《煤中碱金属(钾、钠)含量分级》,煤中钾和钠总量>0.50% 的煤被定义为高碱煤[15];但是从利用角度考虑,灰基下氧化钠含量大于 2% 的煤种属于高钠煤[16]。高钠煤在直接燃用过程中会出现炉内燃烧器区结渣现象和过热器发生超温爆管等问题,从而导致锅炉难以长期稳定运行[17-22],煤中钠含量越高,其结渣沾污倾向越强[23-28]。因此,国内外煤地质学家和煤燃烧、转化领域的学者均高度关注高钠煤的研究。总结近年来关于高钠煤,特别是我国新疆高钠煤的研究成果,针对高钠煤的特殊成因、赋存和影响因素展开论述,并提出符合我国高钠煤含量测定方法、厘清我国高钠煤的富集成因和高钠煤在利用转化中影响钠释放迁移的因素等关键问题,希望能够进一步促进对高钠煤的科学认知和合理利用。

1　煤中钠元素含量测定

研究煤中钠的基础是能够准确测定煤中钠的含量。利用煤灰中 Na_2O 含量和式(1)来折算煤中钠含量是最常见的方法[15,29-32],其他常用方法有低温灰化法[33]、高温灰化法[34]、微波消解法[35]、逐级提取法[36]和氧弹燃烧法[37]等(图 1)。但是运用不同测试方法得到的煤中钠含量的数据有一定的差别,大量的测试数据分析可知,对于新疆高钠煤,逐级提取法和氧弹燃烧法测得钠含量基本一致,国标法测试含量最低,低温灰化法测试结果趋于上述方法之间。同时,逐级提取法在保证总钠含量相对准确的基础上,可以获得各种形态的钠含量。

$$w(Na)_d = 0.742w(Na_2O) \times A_d/100 \quad (1)$$

式中:$w(Na)_d$ 为煤中钠质量分数,%(干燥基);0.742 为氧化钠中钠的质量系数;$w(Na_2O)$ 为煤灰中氧化钠质量分数,%;A_d为煤的干燥基灰分,%。

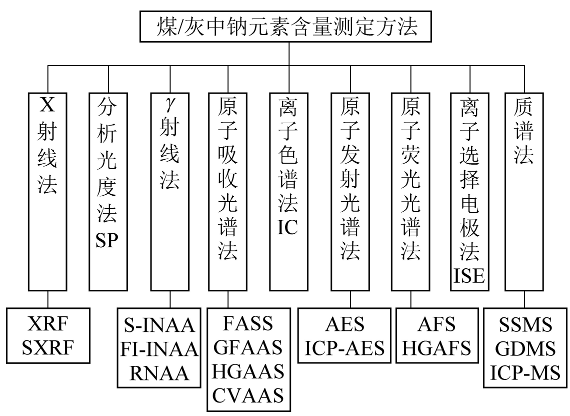

图 1　钠元素测定方法

Fig.1　Determination method of sodium element

1)低温灰化法。低温灰化法有 2 种方法,一种是采用较低温度的灰化温度(温度为 400~550 ℃),另一种采用成灰温度在 100~200 ℃的等离子灰化仪,后者优势在于可有效防止或减少煤中含钠矿物的分解、挥发等。理论上来讲成灰温度越低,获得的

煤中钠含量越高,但低温会造成有机质不能完全灰化,无法保证钠含量的准确性[34]。

2)直接消解法。采用强酸在一定温度程序下直接进行微波消解,进而赶酸、定容,最后通过原子吸收光谱(AAS)或电感耦合原子发射光谱(ICP-AES)测定溶液中的钠浓度,换算成煤中钠的质量百分数。该方法测定的钠损失较少[15],缺点是当煤中钠以矿物态赋存时,由于被包裹的矿物无法完全溶出,会造成测值偏低。

3)逐级提取法。用去离子水、醋酸铵、稀盐酸在室温下对煤样进行提取,测定所得滤液中钠的浓度,可得到水溶态钠、离子交换态钠和碳酸盐结合态钠的含量[38-39];产生的残留物最后消解后即得到不溶态钠含量[37]。此方法是目前测定煤中钠形态的主流研究方法,当研究对象为高钠煤时,测定煤中钠含量约为国标法所测值的 2 倍[40]。

4)氧弹燃烧法。称取一定质量的样品放入装有去离子水的氧弹桶中,通入纯氧并点火灰化,最后对灰样进一步消解、赶酸、定容,测定溶液中的钠浓度,换算成煤中钠的质量分数。本方法是借助氧弹燃烧罐的封闭环境进行燃烧,避免了钠挥发损失[41]。

以上所述的不同测定方法是基于煤中钠赋存状态差异而提出。由于煤中钠在热转化过程中会释放和挥发,且该部分钠对煤燃烧利用时产生的沾污和结渣影响最明显[42-43]。利用高温灰化法会发生钠的逃逸,造成钠含量测值偏低;低温灰化法可以减少钠的逃逸,使其检测值更加接近实际;直接消解法无法消解受晶格约束的钠元素,从而导致部分钠无法检测,此方法不适用含钠矿物较多的煤。当煤中钠的主要赋存状态为水溶态时,逐级提取法可以相对准确地反映煤中钠的含量。氧弹燃烧法的特殊测试条件,更适合热转化后灰中钠含量的测定。有学者研究表明,在煤热转化过程中直接测定气相产物中钠的含量,所得结果更贴近实际[44]。

2 煤中钠元素赋存状态

综合前人研究成果,煤中钠可分为无机钠(如水合离子、含钠矿物等)和有机钠(与羧基或含氮、氧官能团结合)[45]。BENSON 和 HOLM 提出用乙酸铵萃取有机羧酸钠,盐酸萃取存在于含氧、氮官能团上的钠,剩余的钠以铝硅酸盐形式存在[46]。YANG 等[47]用氯化铵代替乙酸铵进行萃取试验,以此排除高溶解性岩盐对钠的影响。此后学者们尝试用水、醋酸铵等溶剂连续提取来分析有机物中钠的赋存状态,最终将钠划分为水溶态、醋酸铵溶态、盐酸溶态和不溶态[48-55]。SUGAWARA 等[56]用其他溶剂(H_2O、CH_3COONH_4、HCl、H_2SO_4和 HF)将钠分为 5 种赋存状态。

中国不同地区煤中钠的赋存状态统计见表 1[40],总体上以水溶态为主。新疆煤中钠含量要远大于我国其他主要产煤区煤中钠含量,水溶态钠含量依旧是主要赋存状态,但占比较高。从我国西部到中部再到东部沿海地区,煤中钠含量呈现出极高值—低值—高值的特征,高钠煤明显集中分布在西部地区。国外高钠煤中含钠矿物主要有黏土矿物、硅酸盐矿物、沸石、岩盐等,芒硝和钠长石较为少见[57]。除此之外还发现少量钠的硫酸盐矿物[32,47,53,58-60],无水芒硝(Na_2SO_4)常以白色块状或粉末状出现在褐煤中[61]。水溶钠溶解于孔隙水中,其含量与煤的孔隙结构有关,煤中孔隙越丰富,含钠物质的总量越大;干燥后会变为有机形式(如—COONa)赋存[9]。

表 1 我国部分地区煤中钠元素的赋存状态[40]

Table 1 Sodium mode of occurrence in coal[40]

采样地		钠含量/($\mu g \cdot g^{-1}$)	赋存状态占比/%			
			水溶态	醋酸铵溶态	盐酸溶态	不溶态
新疆	沙尔湖	8 211	72.0	5.0	2.0	21.0
	淖毛湖	2 113	68.0	6.0	13.0	13.0
	神华准东	3 253	81.5	12.4	1.8	4.3
	五彩湾	—	67.3	28.5	0.6	3.6
	木垒	5 700	42.0	26.0	2.0	10.0
	后峡	6 640	25.0	11.0	5.0	59.0
陕西	神木	572	47.0	37.0	4.0	12.0
内蒙古	准格尔	277	34.0	10.0	16.0	40.0
江苏	徐州	725	42.0	7.0	8.0	43.0
浙江	晓云	920	20.0	3.0	8.0	69.0

3 煤中钠元素的富集成因

煤中钠在成煤作用各阶段都可以形成,包括泥炭中生物碎屑的累积、碎屑沉积物的输入、泥炭孔隙中溶液的沉淀、成煤后期煤孔隙和裂隙溶液中沉淀等,还可以通过地表水淋滤和地下水驱动效应形成。钠的富集分布受多因素地质作用控制,陆源碎屑物质、沉积环境、成煤植物、岩浆热液活动、地下水活动、风化作用等都可以影响煤中钠的丰度[62-63]。学者们从地质作用角度总结了元素富集的主要影响因

素:源岩控制[64]、沉积控制[65-66]、岩浆热液控制[67]、构造控制[63]、地下水控制[63]、火山灰沉降富集[68]和海底喷流[69,70],不同盆地煤中钠含量与上述各种因素的作用及其程度相关。有研究认为,水溶钠主要来自成煤植物和煤吸收的水分[48],以 NaCl 形式存在的钠主要由地下水带入煤层,与水化学类型有关[59];煤中 Na^+、Cl^-不平衡,表明了地下水中离子成分对煤中钠富集影响的复杂性[32]。有机态的钠是进入煤层的水溶钠与有机质长期作用的结果,受温度、pH 条件影响,部分离子可交换形式的钠可与羧基、羟基连接,或以配位形式络合于含氧或含氮官能团[45]。不溶钠在特殊地下水环境(如高温、低 pH 等)条件下随地下水向煤层渗透与其赋存状态有关,随着煤阶的增加,煤有机结构经历的芳构化作用会排出非矿物无机物,导致其含量随煤变质程度提高而降低,进而导致多赋存状态的可溶钠流失,不溶钠堆积残留,并呈现随煤阶增加而增加[32]。

泥炭发育在钠含量高的水体中,或成岩作用阶段高矿化度的地下水与煤层接触均易形成高钠煤。受到海水影响的泥炭沼泽中钠和氯含量较高,RAYMOND发现美国佛罗里达红树林泥炭中钠含量可达 3.69%,泥炭样品灰化后可见岩盐晶体[10];乌克兰西顿巴斯石炭纪煤、德国鲁尔 Frotuna-Garsdorf 褐煤和澳大利亚褐煤在成煤期均因受到海水侵入的影响导致煤中钠含量高[11,65,71]。

成岩作用阶段地下水化学特征和水文地质条件对煤中钠的形成和分布具有重要影响。任德贻等[71-72]认为地下水作用富集型成因的元素与地下水化学性质、水位与煤层的相对关系有关,也和煤系地层性质有关,元素通过淋滤作用带入煤层。保留在煤中的钠无法通过上覆地层的再改造而发生迁变,地下水运移的活跃程度对煤中钠的保存起重要作用[73-74]。例如乌克兰西顿巴斯石炭纪煤在地下水活跃区域的含钠量高于地下水滞流区域[75]。

煤层埋藏深浅也是钠元素富集的重要地质因素。最典型的是澳大利亚维多利亚高钠煤中钠含量随煤层埋深增加而降低。当煤埋深较浅时,上覆岩层的蒸发引力和承压水的静压力使煤中水由下而上运移,可溶性钠在煤层顶部相对富集[76]。煤与围岩化学组成的差异也是造成钠元素富集分布差异的一个重要因素,煤的有机质实际上是作为地下水渗透过程中的离子交换介质存在的,与有机质结合不紧密的离子(如钠离子)的迁移速度要比结合紧密的离子(锗、钼等)要快,因而在煤层的顶部富集[62]。新疆准东煤中钠的空间分布特征和钠的存在形式(氯化钠、硫酸钠以及水合离子钠等)均受到地下水水化学类型、径流特征和煤层的埋藏深度的影响[32]。YANG 等[47]认为,准东高钠煤孔隙丰富,吸附水的能力强,造成煤中水溶钠含量占比高。

4　煤中钠的迁移、分布与其影响因素

4.1　钠在煤利用过程中的迁移规律

煤中钠的迁移一方面与其在煤中的赋存状态相关,另一方面受反应条件的影响。关于钠的释放形式有很多理论和假说,综合有 Na^+、NaCl、NaOH、Na_2SO_4等观点。一般认为煤中钠元素主要以 Na^+形式释放[77-79],主要与 Cl^-和 SO_4^{2-}反应。水溶性钠多以 NaCl 的形式释放,有机钠多以醋酸钠形式挥发,会进一步分解成二氧化碳和钠;小部分有机钠在燃烧过程中被氧化成钠的氧化物,存在于固相残留物中[80-81]。

关于煤中钠在燃烧过程中的不同温度条件下释放规律的研究,认为有机钠从 200 ℃ 开始挥发[54,82],紧接着水溶钠开始释放,部分直接蒸发[83],部分随着水分的蒸发析出至煤表面[84];到 1 000 ℃左右时所有形态的钠释放基本结束[85](图 2)。不同温度下钠的释放形式也有所差异:200~400 ℃时醋酸铵溶态钠含量增加,残渣态钠含量下降[86-87];400~600 ℃时主要以离子态释放,钠释放最快;600~800 ℃时主要以氯化钠形式释放,大于 815 ℃,氯释放完毕,钠主要以硫酸钠形式释放,其冷凝试验表明在 815 ℃以前主要以氯化钠结晶,1 000 ℃则为硫酸钠[24]。当温度低于 1 000 ℃时,煤中水溶态钠会在水分的携带作用下以氯化钠形式析出,当温度高于 1 000 ℃时煤中的部分钠以硫酸盐形式析出,另一部分则会在水的作用下以钠的氢氧化物析出[88]。

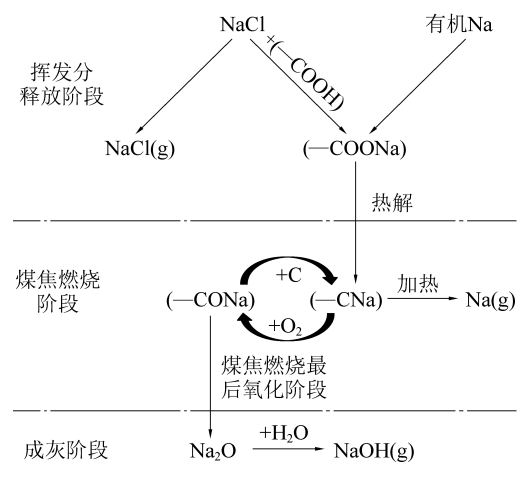

图 2　燃煤过程中钠的析出途径[85]

Fig.2　Sodium precipitation pathway during coal burning[85]

当煤炭在热解时,随着挥发分的大量释放,煤中大部分的水溶性钠会转化成盐酸可溶性钠存在于有机质中[89]。相比于燃烧过程,温度超过 600 ℃时钠才会析出[22],并且随热解温度升高和热解时间的增加,煤中水溶钠比例显著降低,不溶钠比例增大,钠析出至气相中的比例增大[46]。热解过程中以 NaCl 形态存在的钠比以羧酸盐形态存在的有机钠析出容易得多,且热解过程中钠与氯析出比例并不一致[90]。随着热解温度的升高,钠在热解温度为 700~900 ℃集中析出[91]。相比于热解过程,气化过程钠析出率大于热解过程[92];而与燃烧相比,气化时底渣、飞灰残留物中钠的含量更高[93]。煤中钠含量对气化过程沾污结渣有很大的影响,煤中钠含量越高,气化过程结渣现象越明显[94]。此外,气化过程中添加剂的使用及其他矿物质的变化也会显著影响钠的迁移[95]。

4.2 转化产物中钠的分布规律

煤受热后产生的固液气三相中均含有钠,含钠产物还会相互反应,形成多种类的释放途径。例如,挥发至气相的氯化钠会与烟气中其他组分发生反应生成含钠硫酸盐,随后在受热面冷却作用下冷凝沉积[96-98]。基于此,学者们分别从动力学和热力学角度提出了气相钠的硫酸盐的同相生成机理模型[99-103]。在燃烧过程中煤灰中的钠会与其他元素相互作用形成低熔点的矿物质。目前研究一般认为高钠煤灰中的钠主要以钠长石、霞石等形式存在,此外硬石膏、石英、透辉石、赤铁矿、方解石等矿物中可能含钠。其中钠长石熔点较低,铝硅酸盐矿物、霞石、透辉石等会相互反应生成类长石矿物(钙黄长石、镁黄长石)[104-105]。极端情况下,当钠含量超过 20%时,煤灰中的钠元素基本上全部以菱硅钙钠石的形式存在[105]。

4.3 钠迁移转化影响因素

在讨论煤中钠迁移释放规律的同时,学者们也关注控制煤中钠迁移释放的各种因素。通过对钠的释放进行模拟,发现影响钠释放的因素有很多[106-107],大体可以分为外部环境因素和自身性质因素。外部环境因素体现在温度和压力的影响,升温过程中,压力减小,气相中含钠物质会增加。煤自身对钠释放有着至关重要的影响,煤中矿物质的含量和分布,具有吸收钠作用的物质对其燃烧释放有很大影响[108]。例如铝硅酸盐矿物对钠的释放具有明显的抑制作用,当煤中钠含量较低时,氯对钠释放的促进作用不明显[109-110]。煤中方解石则对钠释放起到促进作用,然而形成无水芒硝则对钠释放起到抑制作用[104]。当煤中氯含量升高,氯化钠含量增多,并且氯化钠熔点低易气化,因此气相钠含量会增加[92,111]。除上述主要影响因素外,煤粉的粒径、气氛、反应时间等都会对钠的释放产生影响[112],总之,内外影响因素的多样性和复杂的煤燃烧过程造就钠在燃烧后期的转化方式存在差异。

5 结论与建议

1)煤中钠的赋存状态以无机钠为主,少量为有机钠;新疆煤中钠以水溶态为主;燃烧过程中钠大量进入气相中而不是灰中,会造成煤中钠含量测值偏低,逐级提取法则可以相对准确地反映煤中钠的含量。

2)高钠煤燃烧、热解和气化过程中,钠的迁移和释放特性与其在煤中的赋存状态及热处理条件密切相关。有机钠从 200 ℃挥发,400~600 ℃时主要以离子态释放,600~800 ℃时主要以 NaCl 形式释放,1 000 ℃释放基本结束。热解过程中钠析出的规律与燃烧过程相同,但析出温度要比燃烧温度高;气化过程钠的析出率大于热解和燃烧过程。除了从技术方面的研究,也应从煤中钠的赋存状态和地质成因等的实际情况出发,为脱钠提质提供直观的依据和扎实的理论。

3)多学科交叉研究对科学认识新疆煤中钠的成因、富集和转化机理有重要意义。今后应重点从水文地球化学角度出发,建立钠进入煤层的地下水动力学模型,阐明新疆高钠煤的成因机制,并进一步深入认识高钠煤燃烧过程中钠与其他组分的结合模式及其结渣沾污机理,为提出有针对性的解决方案打下基础。

4)高钠煤在利用加工过程中大比例的钠释放迁移至热转化产物中。高钠煤或许可以作为高附加值产品的优质天然原材料。在目前无法控制煤中钠迁移和富集的情况下,遵循钠迁移富集的基本规律,开发含钠甚至高钠的副产品作为关键领域的核心材料;再者重点关注高钠煤分质转化技术和清洁利用方向,把握煤中钠的富集迁移、转化条件差异所带来的产物性质变化,做到多级利用。

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