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水泥窑协同处置不同类型危险废物时二噁英排放特性研究

潘淑萍,潘荷芳,牟永铭,徐茵茵
(浙江省环境监测中心,浙江 杭州 310015)

摘 要:水泥窑协同处置固废在减量化、无害化和资源化方面具有显著的优势,然而,焚烧过程中不可避免地生成二噁英,而二噁英的排放量可能受固废的种类和处置量等影响。本文系统地研究某4 000 t/d的水泥窑单独处置电镀污泥、有机溶剂、含镍废物和医疗废物时废气中二噁英的排放水平,同时计算其排放因子,并且与已有的研究进行对比分析。结果表明,水泥窑协同处置不同危险废弃物时对二噁英排放的影响较小,所有排放数据均满足国家标准;二噁英主要通过从头合成反应生成,并以高氯代为主;各排放因子与国外水泥窑协同处置的企业的排放水平相当。该技术是一种切实可行的危险废物处置方式。

关 键 词:危险废物;新型干法水泥窑;共处置;烟气;二噁英

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引 言

危险废物是一类特殊的废物类型,具有易燃性、易爆性、腐蚀性、化学反应性、以及对人体潜在的毒性和传染性,对生态环境和人体健康构成了极大地威胁,一旦危害性爆发,将造成长久的,难以恢复的隐患和后果[1~2]。

实现危险废弃物的安全处置和处理,以达到减量化、无害化和资源化的目的,是处置危险废弃物面临的关键问题[3]。目前,普遍采用的处理危险废弃物的方法有安全填埋、焚烧法、热解法、固化处理以及物理、化学和生化处理等[4]。我国相对于国外发达国家在危险废物处置方面起步较晚,主要以填埋和焚烧为主。又因填埋法对地下水等仍然存在潜在的污染,后又逐渐从“填埋”转向“焚烧”。

从现有的焚烧运行情况来看,简单的焚烧过程很有可能使得产生的飞灰和灰渣中残留毒性极高的二噁英,其来源主要来自于两条途径:(1)前驱物合成,即未完全燃烧产物氯酚、氯苯等小分子污染物在飞灰表面合成二噁英;(2)从头合成,即飞灰中的残炭或者炭黑在金属催化剂的催化作用下与氧、氯、氢等反应释放二噁英[5]。对灰渣等进一步采用填埋等二次处理方法,又会造成新的二次污染及增加成本[6]。此外,焚烧系统投资较大,而且国家对环境标准的加大,使得尾气处理成本也在逐步加大,焚烧工艺难以得到大规模普及实施。

新型干法水泥窑焚烧技术是利用水泥回转窑在高温煅烧水泥熟料的同时,焚烧处理危险废物,是一种符合可持续发展战略的新型环保技术。利用新型干法水泥窑焚烧处理生活垃圾和危险废弃物,不仅可以实现废弃物的有效处置,而且可以解决部分燃料来源问题。然而,固体废弃物的焚烧总是不可避免的产生一定量的二噁英,水泥回转窑掺烧不同类型的危险废弃物有可能会对二噁英的生成和排放产生一定的影响[3,7]。

由于分类困难,目前的研究多以掺烧组分复杂的危险废弃物为主,缺乏对处置特定危险废物时的二噁英生成和排放特性的研究。本文研究4 000 t/d水泥生产线在协同处置各类危险废弃物(电镀污泥、有机溶剂、含镍废物、医疗废物)时烟气中二噁英排放浓度和同系物分布规律。

这些数据的提供,不仅能够为处置前预处理工艺设计提供参考,而且能够实现协同处置前对危险废物种类的初步判定,进而对水泥窑协同处置不同种类的固体废物的安全性和可靠性进行评估,有利于相关部门对危险废物集中管理、调配和资源化利用。

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作者简介:潘淑萍(1977-),女,山东青岛人,浙江省环境监测中心高级工程师.
通讯作者:潘荷芳(1965-),女,浙江省环境监测中心教授级高级工程师.

1　材料和方法

1.1　系统介绍

该新型干法回转窑水泥生产线配备五级双系列预热器和分解炉，同时配备 Φ4.8 m×72 m 回转窑。不同危险废弃物收集后运输至暂存车间，大块废弃物需要经过一定的破碎预处理过程通过入窑进料装置方可进入水泥回转窑进行处置。

1.2　试验工况

项目依托原有窑尾烟气除尘系统的同时，在窑尾增设布袋除尘器及脱硝装置，形成电袋复合除尘器、低氮燃烧器 + SNCR 脱硝装置。SNCR 脱硝装置即利用氨水溶液，以雾状喷入水泥窑分解炉处，把烟气中的氮氧化物还原成氮气，从而达到脱硝的目的。窑尾废气处理流程如图 1 所示。监测点位设置在窑尾除尘器出口位置。试验过程中两条水泥生产线的生产负荷约为 100%，新型干法回转窑内物料烧成温度须保证在约 1 450 ℃，且物料在窑中高温下停留时间大于 20 min，窑尾监测点烟气的温度大约为 85 ℃。试验工况如表 1 所示。

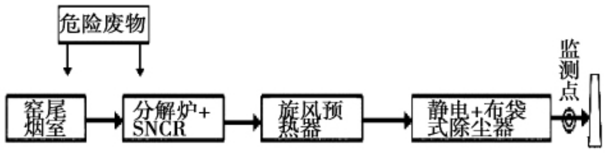

图 1　窑尾废气处理及监测点位示意图

Fig. 1 Schematic diagram of flue gas cleaning system and monitoring points

表 1　水泥窑协同处置不同危险废弃物试验工况

Tab. 1 Experimental conditions in cement kiln when co-processing different hazardous wastes

工况	熟料生产能力/t·d^{-1}	处置废物名称	添加量/t·h^{-1}	烟气量/m^3·h^{-1}
A		空白(无)	0	
B－1		电镀污泥	11.46	
B－2	4 000	有机溶剂	9.17	6.50×10^5
B－3		含镍废物	8.54	
B－4		医疗废物	5.21	

1.3　样品采集

二噁英样品的采集严格按照《HJ77.2－2008 环境空气和废气二噁英类的测定同位素稀释高分辨气相色谱－高分辨质谱法》执行，采样仪器型号为 TCR－5，利用等速采样原理，使烟气分别通过 XAD2/PUF 的吸附阱和冷凝物收集瓶，将烟气中的固相二噁英和气相二噁英吸附，采样结束后用有机溶剂清洗采样管路，收集淋洗液，保存好样品，带回实验室进行下一步的检测分析[8]。二噁英浓度每个周期采样 3 次，计算烟气样品中 17 种有毒二噁英的浓度和对应的毒性当量，并将结果折算至 10% O_2。计算各种污染物的排放因子，即每生产 1 t 熟料排放的污染物总量。

1.4　样品分析

使用加速溶剂提取仪(ASE 300，Dionex)对采集的样品进行萃取[9-10]。萃取条件为：1.03×10^7 Pa，120 ℃，萃取溶剂采用甲苯和丙酮体积比 95∶5 的混合溶液，静态平衡时间为 7 min，萃取循环 3 次，淋洗 70% 体积，吹扫时间为 100 s。在萃取前添加同位素标记的内标物质(Internal Standards IS，EPA0023A，Wellington)。样品萃取液分别经过浓硫酸酸洗，酸碱多层硅胶柱和活性炭分散硅胶柱等净化处理，然后氮吹浓缩，添加进样内标等步骤完成净化处理。最后使用高分辨色谱－高分辨质谱联用仪定性定量分析。分析条件为：60 m×0.25 mm，膜厚未公开的 BPX－DXN(毛细色谱柱的产品编号)，进样口 280 ℃，不分流进样，载气 He 99.999%，流量为 1.2 mL/min。程序升温：130 ℃保持 1min，由 15 ℃/min 至 210 ℃，再由 3 ℃/min 至 310 ℃，然后以 5 ℃/min 至 320 ℃，动态分辨率 >10 000。

2　结果与讨论

2.1　质量控制和质量保证

采样和分析过程中严格执行相关标准，采样内标的回收率为 70% ～90%，净化内标的回收率为 30% ～110%，所有分析结果符合《HJ77.2－2008 环境空气和废气二噁英类的测定同位素稀释高分辨气相色谱－高分辨质谱法》所规定的质量保证要求。

2.2　水泥窑掺烧不同类型危险废弃物对二噁英排放浓度的影响

通过研究不同类型危险废弃物的掺烧对二噁英排放特性的影响，可为综合判断水泥窑协同处置生

活垃圾技术的安全性和环保方面的影响提供必要的依据。经过现场采样及实验室检测分析，烟囱二噁英排放数据如图 2 所示。

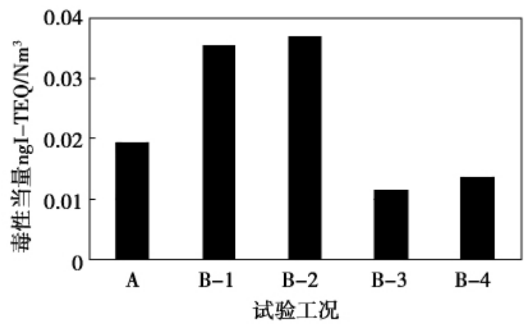

图 2　不同工况下的二噁英排放总毒性当量

Fig. 2 Total I-TEQ from stackunder different operating conditions

如图 2 所示，水泥窑掺烧不同危险废弃物时，烟囱的二噁英平均排放浓度在 0.012 ~ 0.037 ng TEQ/Nm^3之间，低于 0.1 ng TEQ/Nm^3 的排放标准，与文献中日本水泥窑掺烧垃圾二噁英排放浓度 0.096 ngTEQ/Nm^3 水平一致[3]。同一生产线处置不同危废时，二噁英排放量存在显著差异，但是由于样品数量较少，单位时间内处置量不同，并且水泥生产线自身存在扰动，因此并不能说明危险废弃物种类对烟气中二噁英的排放浓度存在影响。

Chen 等人对国内 5 个水泥窑二噁英排放进行了检测分析[7]，发现二噁英排放含量为 2.3 ~ 40 ng/Nm^3，毒性当量为 0.009 3 ~ 0.090 8 ng TEQ/Nm^3。Nguren 等人对越南 3 座水泥厂进行了检测[11]，二噁英排放含量为 0.28 ~ 5.32 ng/Nm^3，毒性当量为 0.033 ~ 0.837 ng TEQ/Nm^3。水泥窑处置单一种类的危险废弃物时废气污染物的浓度均低于排放限值，并不会因为危险废弃物共焚烧而造成污染物排放超标。

分析认为，水泥窑协同处置危险废弃物时对二噁英的排放影响较小的原因主要是由于其本身技术优势决定的，该技术具有以下优点：焚烧温度高、物料停留时间长、良好的湍流、对重金属有一定的固化作用，完全可以保证有机物的完全燃烧和彻底分解。以上优点保证了水泥窑协同处置危险废弃物时焚烧状态稳定，无废渣、废液排出，物料适应性强，建设投资较小，运行成本较低，总体的废气排放量较少，通过烟气净化系统的处理，可以使烟气排放达到国家规定的排放标准[12 ~ 13]。

同时，相关研究也表明，水泥窑内部天然的碱性环境以及生料自身良好的吸附性能均有利于抑制二噁英的生成和排放[14]。

最后，李叶青等人研究认为单独采用静电除尘器有可能提高二噁英的排放量，且可能的原因包括：（1）静电除尘器的运行温度在 200 ~ 300 ℃，刚好位于二噁英的最优合成温度范围；（2）静电除尘器工作过程中的电离环境产生的活性分子，如氢氧根等，对二噁英的生成有促进作用[15]。布袋除尘器相对于静电除尘器，具有初始投资低，除尘效率高，运行费用低等优点，布袋除尘器可以有效从源头控制大气中二噁英的主要来源。在该系统中，由于在窑尾增设布袋除尘器，形成电袋复合除尘器也有利于二噁英等的进一步脱除。

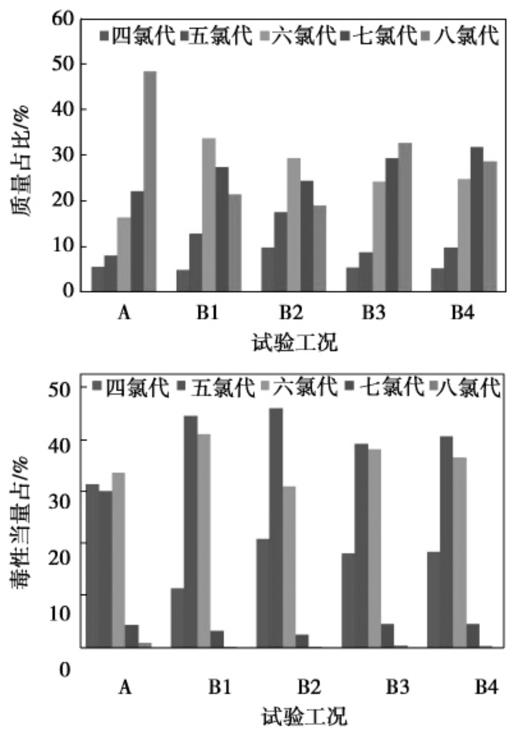

图 3　二噁英排放特性（质量和毒性当量占比）

Fig. 3 Characteristic of PCDD/Fs emission from stack (mass - and I - TEQ ratio)

2.3　水泥窑掺烧不同类型危险废弃物对二噁英指纹特性的影响

掺烧不同类型的危险废弃物在二噁英质量分布上存在共性。二噁英主要以生成高氯代二噁英为

主，占到了 72.6% ~95.6%，平均氯化度则在 6.25 ~7.13 之间；主要的生成物为 OCDD 和 1234678 - HpCDF；普遍认为 PCDD/Fs 的生成机理包括高温气相催化反应以及低温催化合成反应，而后者又包括前驱物异相催化生成和从头合成反应。研究发现，PCDD 和 PCDF 的生成机理是不同的，在典型的燃烧过程中，由从头再合成生成 PCDD/Fs 的反应途径中，其 PCDFs/PCDDs 的比值在 1.6 左右，而在由前驱物异相反应生成 PCDD/Fs 的反应途径中，其比值则远小于 1。不同工况下 PCDFs/PCDDs 的比值范围在 1.17 ~2.21 之间变化，这从侧面说明水泥窑协同处置危险废物过程中，从头再合成反应在二噁英生成途径中占主导。

2.4　排放因子分析

Liske 等人早在 1996 年就从水泥生料中检测到了二噁英的存在[16]。Dyke 等人对水泥窑的粉尘做了检测分析[17]，发现英国水泥窑粉尘的二噁英含量水平为 0.001 ~30 ng TEQ/kg，德国水泥窑粉尘二噁英含量水平为 1.0 ~40.0 ng TEQ/kg，瑞士为 0.03 ng TEQ/kg。在排放因子方面，欧洲为 0.15 μg TEQ/ton[18]，西班牙为 0.014 μg TEQ/t[19~20]，英国为 0.025 - 1.2 μgTEQ/t[21]。

根据危险废弃物掺烧量、烟气量及相应的二噁英排放量，也可计算不同工况下生产单位重量的熟料时二噁英的排放情况，本文计算二噁英排放因子如下：

$$F = C_f \times V_f / M$$

式中：F—二噁英排放因子；C_f—布袋过滤后烟气中二噁英浓度，mg/m^3；V_f—烟气流量，m^2/s；M—二噁英采集过程中熟料的产量。

根据熟料生产量、烟气量及相应的二噁英排放量，也可计算得到相应的排放因子，如图 4 所示。值得注意的是，二噁英在水泥窑系统中是一个沉降过程，对于 5 000 t/d 的生产线每年会沉降 669 ~1 325 mg毒性当量的二噁英[5]。因此，大力推广水泥窑协同处置固废有利于减少环境中二噁英的存在量。

可以发现，水泥窑协同处置固废前二噁英的排放因子为 0.073 μg TEQ/t 熟料。掺烧不同种类危险废弃物会导致二噁英排放因子发生不同程度的变化，尤以共处置有机溶剂时的排放因子最高，达到了 0.146 μg TEQ/t 熟料，同时水泥窑协同处置含镍废物和医疗废物时排放因子相比于空白试验均有所下降，表明水泥窑协同处置危险废弃物对二噁英的排放因子影响较小。对于相同规模的水泥生产线而言，排放因子大小顺序为：有机溶剂 > 电镀污泥 > 医疗废物 > 含镍废物。

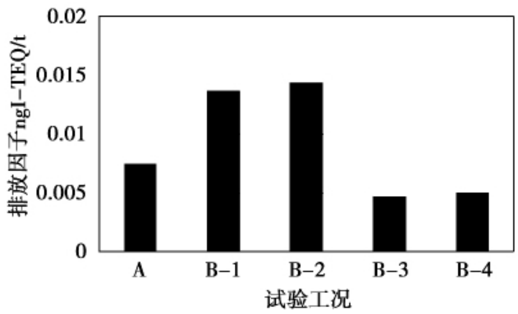

图 4　水泥窑共处置不同危废时二噁英的排放因子

Fig. 4 Emission factors in cement kiln when co-processing different hazardous wastes

3　结　论

水泥窑因其温度高、停留时间长、天然的碱性环境等优势成为处置固体废物的理想方式，不论从质量浓度还是毒性当量方面，协同处置不同种类的固体并未对二噁英的排放造成明显的负面影响：

（1）水泥窑协同处置固废前二噁英毒性当量排放浓度为 0.020 ng TEQ/Nm^3。水泥回转窑分别掺烧电镀污泥、有机溶剂、含镍废物和医疗废物时窑尾烟气中二噁英的排放浓度分别为 0.035、0.037、0.012和 0.013 ng TEQ/Nm^3，远低于 0.1 ng TEQ/Nm^3的排放标准。通过分析认为不同种类危险废弃物的掺烧对水泥窑二噁英的排放特性影响不明显。

（2）水泥窑掺烧不同种类危险废弃物时二噁英同系物研究表明掺烧不同类型的危险废弃物在二噁英质量分布上存在共性，其中二噁英主要以生成高氯代同分异构体为主，同时从头再合成反应可能是水泥窑协同处置危废时二噁英合成的主要途径。

（3）水泥窑协同处置固废前二噁英的排放因子为 0.073 μg TEQ/t 熟料，掺烧电镀污泥、有机溶剂、

含镍废物和医疗废物时窑尾烟气中二噁英的排放因子分别为 0.138、0.146、0.047 **和** 0.067 μg TEQ/t **熟料，与国外水泥窑协同处置的企业的排放水平相当。**

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（单丽华　编辑）

煤灰对铁基载氧体理化特性的影响 = Effect of Coal Ash on the Physicochemical Property of Iron-based Oxygen Carrier 刊，汉

ZHANG Shuai (ShenhuaGuohua (Beijing) Electric Power Research Institute Co. , Ltd. , Beijing China, Post Code: 100025), XIAO Rui (Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, Southeast University, Nanjing, China, Post Code: 210096) // Journal of Engineering for Thermal Energy & Power. -2016, 31(12). - 81 ~ 86

Abstract: The coal ashes derived from three typical Chinese coals (lignite, bitumen, and anthracite) were prepared and used to investigate the interaction between coal ash and iron-based oxygen carrier in a thermogravmetric analyzer, and the effect of coal ash on the reactivity, recyclability and physical structure of iron-based oxygen carrier was evaluated. The results showed that all three coal ashes could enhance the fuel gas conversion in the reduction period, in which anthracite ash behaved the best with the highest fuel conversion achieved. Compared to the stable cyclic performance of iron-based oxygen carrier without coal ash presented, the recyclability of iron-based oxygen carrier showed different degrees of degradation after three coal ashes were added. The reason could be ascribed to the sintering on the surface of oxygen carrier particles, which was mainly caused by the ash deposition on the iron-based oxygen carrier. Of all the prepared coal ashes, anthracite ash was showed to have the minimal impact on the physical structure of iron-based oxygen carrier. **Key words:** chemical-looping combustion, Chinese coal, coal ash, iron-based oxygen carrier, interaction

基于最小二乘支持向量机的煤耗率软测量 = Soft Sensing of Coal Consumption Rate based on Least Squares Support Vector Machine 刊，汉

DU Hai-ling (Shanxi University, Taiyuan, Shanxi, China, Post Code: 030000) // Journal of Engineering for Thermal Energy & Power. -2016, 31(12). - 87 - 91

To measure coal consumpti on rate accurately on the fly and improve the economy of boiler unit, this article used the method of least squares support vector machine (LSSVM) to build a soft measurement model for the coal consumption rate of a sub-critical boiler, and it was validated by the unit operation data of one year long. The comparison showed the maximum relative error of the soft measurement is 5.7% and it meets the industrial precision requirement. This paper also introduced a correction method for the on-line measurement. The results showed that with the on-line correction the maximum relative error becomes 2.2%, further improving the accuracy of soft measurement, and it can be applied in the process of electric power generation. **Key words:** soft sensing, least squares support vector machine, coal consumption rate, correction factor

水泥窑协同处置不同类型危险废物时二噁英排放特性研究 = Emission Characteristics of PCDD/Fs from

Cement Kiln when Co-processing Various Hazardous Wastes [刊,汉] PAN Shu-ping, PAN He-fang, MOU Yong-ming, XU Yin-yin (Zhejiang Province Environmental Monitoring Centre, Hangzhou, Zhejiang, China, Post Code: 310015) // Journal of Engineering for Thermal Energy & Power. -2016, 31(12). - 92 ~ 96

Co-processing solid wastes with cement kiln has the advantages of volume reduction, pollution reduction and resource utilization. However, PCDD/Fs (polychlorinateddibenzo-p-dioxins and dibenzofurans) are inevitably formed during the incineration process. The emission level may be influenced by the type of wastes and the amount of disposal, etc. This study investigated the emission levels and factors of cement production line (4000t/d) when co-processing various hazardous wastes (including electroplating sludge, organic solvent, nickel-contained waste, and medical waste). The results were compared with previous study and showed that co-processing various hazardous wastes in different scales of cement production has little effect on the emission levels. All the emission data meet the current national standards. The PCDD/Fs are mainly formed by de novo pathway and exist in highly chlorinated PCDD/Fs. The emission factors are comparable to that of foreign cement kilns. This technology is demonstrated to be a feasible way for hazardous waste disposal. **Key words:** hazardous wastes, new type dry cement kiln, co-processing, flue gas, dioxins

联合循环余热锅炉汽包强度可靠性设计计算 = Design Calculation on the Strength Reliability of Waste Heat Boiler Steam Drum for a Combined Cycle [刊,汉] ZHENG Xin-wei (College of Power and Engineering, Harbin Engineering University, Harbin, Heilongjiang, China, Post Code: 150001), MIAO An-li (Wuxi Subsection of No. 703 Research Institute, CSIC, Wuxin, Jiangsu, China, Post Code: 214151), DU Xiao-jian (No. 703 Research Institute, CSIC, Harbin, Heilongjiang, China, Post Code: 150078), WANG Guang-yan (Power Department, Harbin Electric Power Vocational Technology College, Harbin, Heilongjiang, China, Post Code: 150030) // Journal of Engineering for Thermal Energy & Power. -2016, 31(12). - 97 ~ 102

Boiler strength reliability is of significant importance to improve the boiler quality and market competitiveness. In this paper, the calculation on the strength reliability of steam drum in practical engineering was introduced, and some universally applicable determination methods on the design parameters of important random variables were put forward, and followed by the strength reliability calculation methods on the head of the straight section and steam drum circumferential weld seam, and the determination of the target reliability of strength for the whole boiler and every pressure part. Increasing the difference between nominal thickness and design thickness is conducive to improve and ensure the strength reliability. The calculation methods of strength reliability presented in this paper are suitable for various boiler drums. This study is believed to extend the existing researches, and set a new requirement of strength reliability for boiler pressure parts, which are currently calculated through the numerical relationship of