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基于多元线性回归的污泥水热炭燃料特性研究

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摘　要

水热处理是一种高能效的污泥绿色预处理方法，可显著改善污泥脱水性能。掌握水热处理对污泥燃料特性的影响规律是顺利实现其后续热化学转化的前提。利用皮尔逊相关性分析评估原始污泥理化性质和水热处理条件与水热炭理化性质之间的线性相关关系。结果表明，水热炭中氢、氮、氧和挥发分含量与水热温度具有较强的负相关关系，而灰分和固定碳含量与水热温度具有较强的正相关关系。高位热值主要受原始污泥中碳、氢和灰分含量的影响，与原始污泥中碳、氢含量呈极强的正相关关系，而与灰分含量呈极强的负相关关系。高灰分含量会影响水热炭中碳含量变化，转化为干燥无灰基后，碳含量随着水热温度升高而增加。但较高的水热温度不利于水热炭收率增加。相比于水热温度，反应时间的影响较弱，上述结论与实验结果高度一致。根据方差分析建立包含具有统计意义变量的回归模型，模型精度较高(*Adj.*R^2 > 0.8, *RMSE* < 5%)，但对水热炭收率和含水率的预测精度略低。研究结果对水热炭燃料特性评估和水热处理条件优化具有指导意义。

关键词

污水污泥，水热处理，水热炭，燃料特性，多元线性回归

Research on Fuel Properties of Sewage Sludge Derived Hydrochar Based on Multiple Linear Regression

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Abstract

Hydrothermal (HT) treatment is an energy-efficient and green method for sewage sludge (SS) pretreatment, which can significantly improve the dewatering performance of SS. Mastering the effect of HT treatment on fuel properties of SS is the premise of its subsequent thermochemical conversion. Pearson analysis was adopted to evaluate the linear correlation among feedstock, HT conditions and hydrochar properties. Results showed that H, N, O and volatile matter (VM) contents in the hydrochar had a strong negative correlation with HT temperature, while ash and fixed carbon (FC) contents had a strong positive correlation with HT temperature. Higher heating value (HHV) of hydrochar was mainly affected by C, H and ash contents of raw SS, which had a strong positive correlation with C and H contents, but a negative correlation with ash content. High ash content of SS would overlap the variation of C content in the hydrochar, and it was found that C content augmented substantially with increasing HT temperature after transformation into dry and ash free (daf) basis, while the hydrochar yield decreased greatly. Compared with HT temperature, the influence of the reaction time was relatively weak. Good agreement has been observed between the Pearson analysis and experimental results. Subsequently, MLR models including statistically significant variables were developed based on *ANOVA* analysis, suggesting that these models fitted the data associated with hydrochar properties well in terms of $Adj.R^2$ > 0.8 and *RMSE* < 5%, while the prediction of yield and moisture content was acceptable. The research results can provide the guidance to evaluate fuel properties of hydrochar and optimize HT conditions.

Keywords

Sewage Sludge, Hydrothermal Treatment, Hydrochar, Fuel Properties, Multiple Linear Regression

1. 引言

城市污水污泥(以下简称“污泥”)是污水处理过程产生的副产物[1]。随着城市化和工业化进程的加快，我国污泥产量逐年提升，年均增长率达 10.7%。其中，2019 年湿污泥产量达到 6325 万吨[2]。因污泥中含有病原体、重金属和持久性有机污染物等有害物质，若处置不当，易引起二次污染[3]。污泥堆肥已逐渐受限，而卫生填埋也因土地资源稀缺而逐渐减少[4]。同时，污泥中含有大量有机物，可通过厌氧消化或者焚烧、热解及气化等热化学方法将其转化为能源或燃料[5]。为维持污泥自持燃烧，陈等[6]认为污泥入炉前含水率需达到 65%。由于污泥脱水性能非常差，传统机械脱水方式只能将含水率降到 80%左右[4]。而污泥热干化能耗高，同时还会产生 VOCs (挥发性有机物)和 NH_3、H_2S 等污染物[7]。Kim 等[8]认为整个热解处理过程的主要能量输入来自污泥干燥，约占 65%~75%。

水热处理是指在一定温度及自饱和压力条件下，将污泥转化为含碳固体产物水热炭、水热液和少量气相产物(主要为 CO_2)的过程[9] [10]。该过程经历了水解、脱水、脱羧、聚合和芳构化等复杂的化学反应[11]。相比于原始污泥，脱水和干燥性能均得到显著改善[12]。据王等[13]报道，初始含水率为 87%的剩余污泥经水热处理后，通过机械脱水可将含水率有效降至 27%。此外，水热炭 H/C 和 O/C 原子比接近褐煤[14]，具有作为清洁固体燃料使用的前景[15]。

近年来，污泥水热处理研究受到国内外学者广泛关注，同时因水热处理条件和污泥来源等差异导致水热炭理化性质变化较大。而且，实验过程消耗大量时间和人力成本，实验数据易受环境因素与人为因素影响[16]。因此，从有限的实验数据中挖掘、分析并建立合适的预测模型可为未来实验设计提供指导。

基于多元线性回归(MLR)、多元非线性回归(MNLR) (包括回归树和随机森林)和机器学习(ML)等模型对水热炭理化性质的研究较多[16]-[22]。Vallejo 等[17]和李等[19] [21]分别借助多元线性回归模型和多元非线性回归模型对生物质水热炭中碳含量、高位热值和收率进行预测，同时确定了各自的主要影响因素。他们建立的模型中包含了不同来源的生物质，如 Vallejo 等搜集了 70 种生物质数据来建模，而李等[19]搜集的生物质数据也高达 985 组，由此能够保证所建立模型的通用性，但精度较低，李等[19]模型的 $Adj.R^2$ (调整后的决定系数)仅在 0.63~0.79 范围。污泥主要由蛋白质、多糖和脂质等有机物组成[4]，与木质纤维素类生物质组成完全不同，且在水热处理过程中会经历不同的反应途径，因此有必要单独考虑污泥。此外，各学者的研究重点也不尽相同，他们分别针对不同的水热炭理化性质建立预测模型。穆等[16]采用 ANN (人工神经网络)模型并结合 PSO (粒子群优化)算法对固体废弃物水热炭 H/C 和 O/C 摩尔比进行预测，以此来反映水热处理过程中脱水和脱羧基强度。Djandja 等[18]基于 ANN 模型对污泥水热炭中氮含量进行相关性分析，并建立预测模型，发现原始污泥氮含量是最主要的影响因素，其次为挥发分产率和水热温度。郑等[22]基于多元回归方法对污泥水热炭工业分析和元素分析进行预测，除氢元素外，模型整体精度较高，$Adj.R^2 > 0.8$，$RMSE$ (均方根误差) < 3%。

本文较为全面地考虑了水热炭的工业分析、元素分析、高位热值(HHV)和含水率(MC)等理化性质以及水热炭收率(Yield)，并利用皮尔逊相关性分析探究原始污泥理化性质和水热处理条件对水热炭理化性质与水热炭收率的影响，同时基于方差分析结果建立包含具有统计学意义变量的回归模型，研究结果可为水热炭燃料特性评估和水热处理条件优化提供指导。

2. 方法

2.1. 数据收集

数据收集时，以污泥(sewage sludge)、水热处理(hydrothermal treatment)、水热炭化(hydrothermal carbonization)、水热炭(hydrochar)等作为关键词，利用 ScienceDirect、Web of Knowledge 和 Google Scholar 等进行文献检索，收集了从 2013 年到 2022 年共 27 篇相关文献[13] [14] [15] [23]-[46]，提取包括污泥理化性质(初始含水率、工业分析、元素分析)、实验参数(水热温度、反应时间)、水热炭理化性质(含水率、工业分析、元素分析)和水热炭收率等数据，共计 114 组。同时统一污泥及其水热炭工业分析和元素分析数据的分析基准为干燥基，另外，部分数据通过 GetData 软件从文献图片中获取。相关的数据收集情况如图 1 所示。

2.2. 相关性分析

皮尔逊相关系数(r)常用来衡量两变量之间的线性相关关系，可根据协方差和标准差计算得到，如式(1)所示。计算值范围为−1~1，越接近−1 和 1，表明相关性越强，正值代表正相关性，负值代表负相关性，0 代表无相关关系。

$$P(X,Y)=\frac{\operatorname{cov}(X,Y)}{\sigma_x\sigma_y} \tag{1}$$

式(1)中，$P(X,Y)$ 是变量 X 和 Y 之间的皮尔逊相关系数，$\operatorname{cov}(X,Y)$ 是协方差，σ_x，σ_y 分别是各自的标准差。

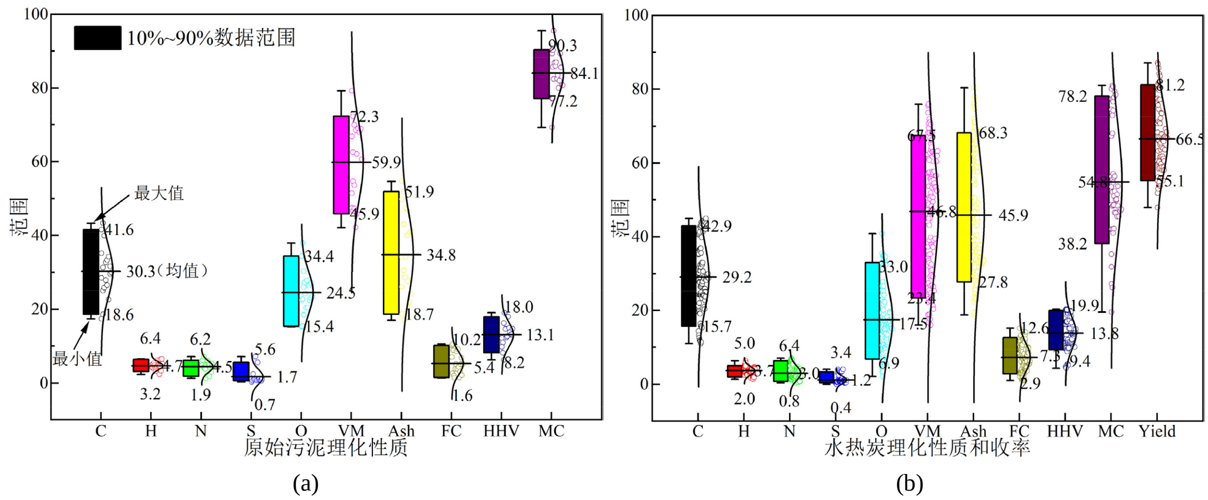


Figure 1. Overview of collected data: (a) Raw SS, (b) hydrochar
图 1. 数据收集情况：(a) 原始污泥，(b) 水热炭

2.3. 多元线性回归模型

回归分析常用于预测，目的是构建一个可以根据自变量的输入来预测因变量的数学模型[19]。建模时，随机地选择 80%的数据用于训练，而剩余的 20%数据则用于测试[47]。利用 SPSS 软件中多元线性回归模块向后消除程序，获得了包含具有统计意义变量($p < 0.05$)的回归方程，回归方程形式如式(2)所示[48]：

$$Y_i = B_0 + B_1 X_1 + B_2 X_2 + \cdots + B_n X_n + \varepsilon \tag{2}$$

式(2)中，Y_i 为因变量，代表水热炭理化性质和水热炭收率；$X_1, X_2, \cdots, X_n$ 为自变量，代表原始污泥理化性质和实验参数；B_0 为截距，$B_1, B_2, \cdots, B_n$ 为相应自变量的回归系数，$B_1, B_2, \cdots, B_n$ 是根据普通最小二乘法(OLS)使残差平方和(*RSS*)最小化的解，残差平方和如式(3)所示[49]，ε 为随机误差。

$$RSS = \sum_{i=1}^{n} \hat{\varepsilon}_i^{\,2} = \sum_{i=1}^{n} \left(Y_i - \hat{Y}_i\right)^2 \tag{3}$$

其中，$\hat{\varepsilon}_i$ 为模型的残差；$\hat{Y}_i$ 为因变量的预测值。

2.4. 模型评估

调整后的决定系数($Adj.R^2$)和均方根误差(*RMSE*)等指标将用于评估模型。其中，调整后的决定系数用于评估模型的拟合优度，它是对决定系数(R^2)的修正，因考虑到每个模型中解释变量的数量，能够更为精确地反映每个模型的拟合优度[19]。均方根误差是衡量预测值和实验值之间的偏差情况，可根据式(4)计算得到[50]。

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} \left(Y_i - \hat{Y}_i\right)^2}{n}} \tag{4}$$

3. 结果与讨论

3.1. 相关性分析

皮尔逊相关性分析可评估自变量(原始污泥理化性质和实验参数)与因变量(水热炭理化性质和水热炭收率)之间的线性相关关系，结果如图 2 所示。水热炭元素分析、工业分析和高位热值受原始污泥理化性

质影响非常大，实验参数的影响与之相比较弱，但也可以发现水热炭中氢、氮、氧和挥发分含量与水热温度具有较强的负相关关系，而灰分和固定碳含量与水热温度具有较强的正相关关系。原始污泥水热处理过程发生的脱水反应导致氢含量降低[11]，傅里叶变换红外光谱(FTIR)分析表明水热炭中-OH 伸缩振动强度会随着水热温度升高而减弱[35]。氮含量降低是因为原始污泥中无机氮和蛋白质在 300℃以下发生水解和脱氨反应，形成铵根离子溶解于水热液中[22]。水热处理还会破坏有机质中含氧官能团(-OH 和-COOH) [22]，使氧含量降低，水热炭 FTIR 分析观察到酮和酰胺基团中 C=O 伸缩振动和-COOH 中 C=O 不对称伸缩表明水热处理过程中发生了脱羧反应[24] [51]，同时水热液中溶解性化学需氧量(SCOD)随着水热温度升高而不断增加[13]，表明水热炭挥发分产率降低。原始污泥挥发分产率因水热处理而降低，矿物质则继续保留，水热炭灰分含量因此而增加。固定碳含量增加是因为高温(大于 240℃)促进聚合反应的进行[24]。与王和李等[13] [52]实验结论一致，高位热值与原始污泥中碳、氢和挥发分含量呈极强的正相关关系，而与灰分呈极强的负相关关系。水热炭收率与水热温度呈较强的负相关关系，这是因为高温会促进原始污泥中有机物的水解[27]，大多数研究[14] [15] [23] [27] [33] [35]均支持这一结论。相较于水热温度，反应时间的影响较弱。

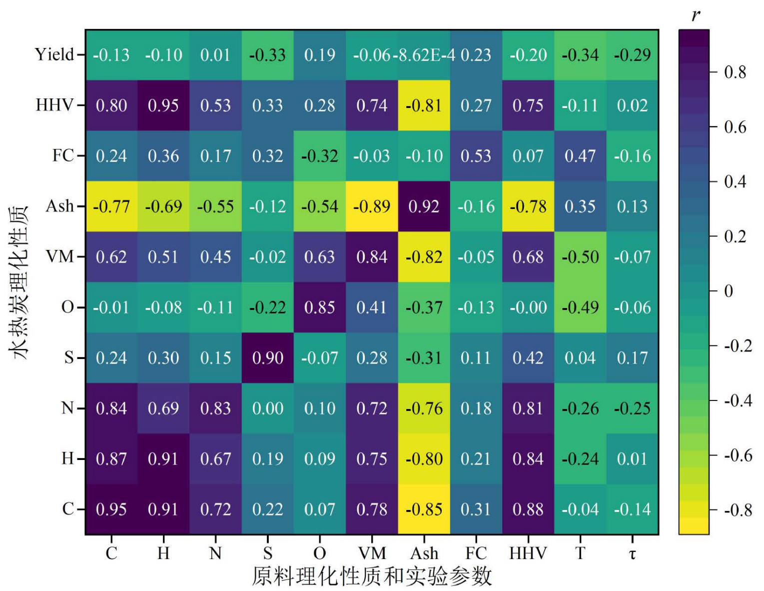


Figure 2. Matrix graph of Pearson correlation coefficient
图 2. 皮尔逊相关系数矩阵图

3.2. 方差分析(*ANOVA*)

方差分析可用于评估原始污泥理化性质和实验参数对水热炭理化性质和水热炭收率是否具有显著影响。一般认为，如果 $p < 0.05$，表明此自变量对因变量具有显著影响，具有统计学意义[19]。根据方差分析，可以选择具有统计学意义的变量建立预测模型，方差分析结果如图 3 所示。

以干燥基进行分析时，*t* 检验结果表明水热温度对水热炭中碳含量没有显著影响，这与实验结果矛盾。王、高和 Areeprasert 等[13] [23] [24]研究表明较高的水热温度可以促进水热炭中碳含量增加；然而，林和王等[28] [30]则发现水热炭中碳含量随着水热温度升高而降低。深入分析数据后发现污泥高灰分含量会影响水热炭中碳含量变化。于是，将分析基准转化为干燥无灰基以剔除灰分的影响，并发现水热温度对水热炭中碳含量具有促进作用，且 *t* 检验结果亦表明水热温度对水热炭中碳含量具有显著影响。

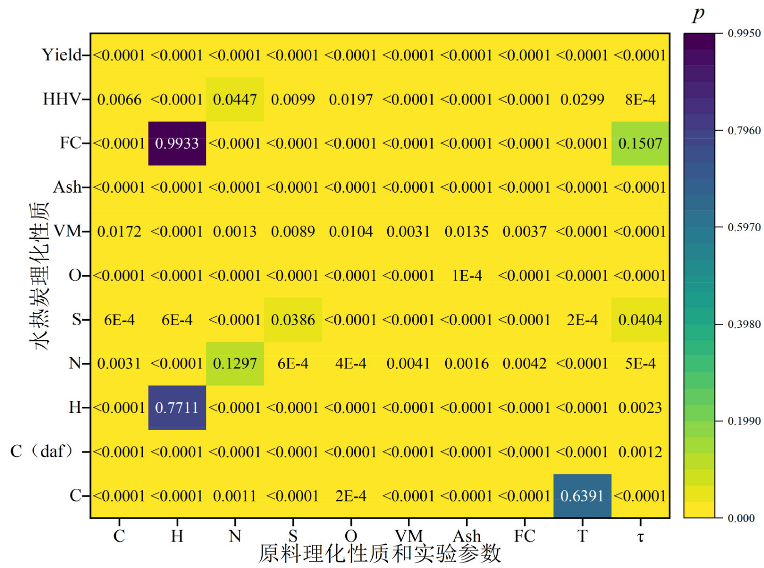


Figure 3. *t*-test results of physiochemical characteristics and yield of hydrochar
图 3. 水热炭理化性质和收率 *t* 检验结果

除了反应时间对水热炭中固定碳含量没有显著影响，实验参数对其它元素分析和工业分析都表现出显著影响。水热处理涉及非常复杂的化学反应，且受实验参数的影响比较大。污泥中多糖在水热处理过程中首先会被水解生成葡萄糖或果糖等单糖，然后分子内脱水转化为 2-糠醛和 5-羟甲基糠醛(5-HMF)等中间体，此外，污泥中蛋白质还会被水解生成氨基酸，并与还原糖通过美拉德反应生成含氮杂环，2-糠醛、5-羟甲基糠醛和含氮杂环在液相中会通过缩合、聚合和芳构化等一系列化学反应形成水热炭[15]。温度的关键作用是提供足够的能量来分解大分子有机物，从而使具有高活性的化学键断裂和重组，而污泥中多糖、蛋白质和脂质等大分子有机物分解的难易程度各不相同，受温度影响比较大[4]。Ruyter [53]建立的水热处理半经验模型表明温度和时间具有一定的等效性，可在有效的温度范围内通过调整反应时间达到相似的炭化水平，因此反应时间对水热炭理化性质也具有一定的影响。此外，原始污泥理化性质对水热炭元素分析和工业分析的影响也非常大，但原始污泥中氢含量对水热炭中氢含量和固定碳含量未表现出显著影响。相比于原始污泥，水热炭工业分析和元素分析发生显著变化，正如皮尔逊相关性分析，挥发分、氢、氮和氧含量会因水热处理而降低，而灰分、固定碳和碳含量会增加。

水热炭热值和收率的 *t* 检验结果显示，原始污泥的理化性质和实验参数都表现出显著影响，表明原始污泥的工业分析、元素分析和水热处理条件都会影响水热炭热值和收率，与报道的实验结果一致。

与其它因变量不同，水热炭含水率建模时只考虑原始污泥的初始含水率和实验参数，但是拟合优度较差，纳入它们的平方项和交互项后，拟合优度提升，如表 2 所示，$Adj.R^2 = 0.9240$，$RMSE = 3.71\%$。

3.3. 回归模型建立与验证

Table 1. Regression equations of physiochemical characteristics and yield of hydrochar
表 1. 水热炭理化性质和收率的回归方程

因变量	回归方程
C_{hc} (%, *daf*)	$C_{hc} = 18C + 21.42H + 14.08N + 15.63S + 16.14O - 83.63VM - 66.65Ash - 83.34FC + 0.084T + 0.015\tau + 6683$
H_{hc} (%, *db*)	$H_{hc} = -0.77C - 0.68N - 0.76S - 0.79O + 4.96VM + 4.18Ash + 4.96FC - 0.0062T - 0.0007\tau - 416.6$
N_{hc} (%, *db*)	$N_{hc} = -0.42C - 1.04H - 0.56S - 0.59O - 6.5VM - 7.09Ash - 6.48FC - 0.015T - 0.002\tau + 709.8$

Continued

S_{hc} (%, *db*)	$S_{hc} = -0.29C - 0.38H - 0.41N + 0.18S - 0.36O - 5.29VM - 5.63Ash - 5.21FC - 0.002T - 0.0003\tau + 563.1$
O_{hc} (%, *db*)	$O_{hc} = -8.03C - 10.51H - 7.4N - 7.96S - 6.78O + 42.85VM + 34.86Ash + 42.92FC - 0.048T - 0.0093\tau - 3479$
VM_{hc} (%, *db*)	$VM_{hc} = -2.73C - 6.91H - 3.91N - 3.17S - 2.92O + 30.52VM + 26.21Ash + 29.81FC - 0.12T - 0.022\tau - 2617$
Ash_{hc} (%, *db*)	$Ash_{hc} = 9.01C + 11.43H + 7.81N + 8.79S + 8.98O - 76.41VM - 66.4Ash - 76.58FC + 0.087T + 0.018\tau + 6726$
FC_{hc} (%, *db*)	$FC_{hc} = -3.66C - 2.52N - 2.92S - 3.33O + 32.67VM + 29.51Ash + 33.5FC + 0.033T - 2953$
HHV_{hc} (MJ/kg, *db*)	$HHV_{hc} = 1.48C + 3.59H + 1.03N + 1.39S + 1.24O + 19.12VM + 20.31Ash + 18.79FC - 0.006T - 0.003\tau - 2035$
$Yield_{hc}$ (%)	$Yield_{hc} = -17.89C - 26.36H - 14.91N - 16.57S - 16.76O - 18.52VM - 35.3Ash - 15.8FC - 0.1T + 0.015\tau + 3666$
MC_{hc} (%)	$MC_{hc} = 129.9M_0 + 4.54T + 3.35\tau - 0.7M_0^2 + 0.0016T^2 + 0.00005\tau^2 - 0.061M_0T - 0.041M_0\tau - 0.017T\tau + 0.0002M_0T\tau - 5826$

水热炭理化性质和收率的多元线性回归方程如表 1 所示，水热炭中碳含量从干燥无灰基转化为干燥基可参考式(5)。

$$C_{hc}(\%, db) = C_{hc}(\%, daf) \times \frac{100 - Ash_{hc}(\%, db)}{100} \tag{5}$$

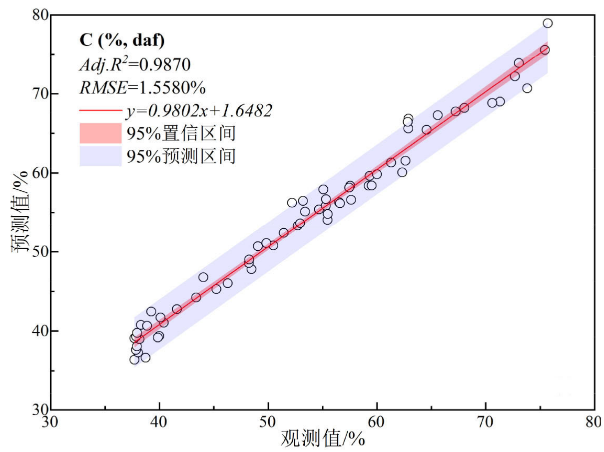
$Adj.R^2$ 和 *RMSE* 可以反映回归模型的精度。此外，比较拟合直线与对角线 $y = x$ 的偏差和预测区间的宽度也能反映所建立模型的精度。如图 4 和表 2 所示，回归模型对水热炭工业分析和元素分析预测精度非常高，$Adj.R^2 > 0.9$，$RMSE < 2\%$，均高于郑等[22]报道的结果。水热炭高位热值、收率和含水率的预测精度相对略低。在整理收集的数据时，发现高位热值大多数是相关仪器测试的结果，也有根据不同的经验公式计算得到[15] [35] [44]，数据来源对高位热值的预测精度产生了一定影响。此外，水热炭收率可能会因脱水方式不同而造成偏差，比如机械脱水会因残留而造成收率偏低，而真空抽滤则在此过程的损失相对较少。水热炭含水率也会因不同的脱水方式而造成偏差，且机械脱水中不同的压力也会导致脱水程度不同。但总体而言，$Adj.R^2 > 0.8$，$RMSE < 4\%$，预测精度较好，建立的水热炭理化性质和水热炭收率回归模型能够有效地预测水热炭燃料特性，为后续热化学转化提供指导。回归模型的验证情况如图 5 所示，可以发现回归模型的验证结果略差于建模，但相差较小，除了含水率，其他因变量 $Adj.R^2 > 0.8$，$RMSE < 5\%$，表明建立的水热炭理化性质和收率回归模型可靠性好。水热炭含水率的验证结果为 $Adj.R^2 = 0.7943$ 和 $RMSE = 6.48\%$。因只有部分文献报道了水热炭收率和含水率等数据，用于建模和验证的数据点比其它因变量少，根据式(4)，*RMSE* 与数据点数量成反比，因而计算出的 *RMSE* 略大，此外，因为水热炭含水率的建模数据点较少，只有在较窄的自变量范围内才能满足其回归模型，当验证数据超过这个区间，会产生较大的偏差。所有回归模型的评估结果汇总于表 2。

Table 2. Summary of MLR model evaluation indicators
表 2. MLR 模型评估指标

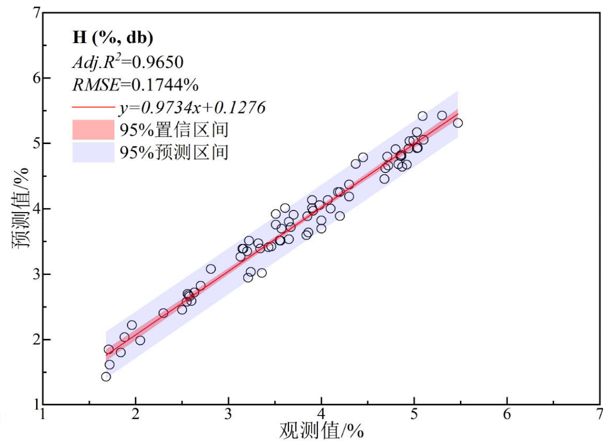
因变量	训练数据集			测试数据集		
	$Adj.R^2$	*RMSE*/%	数据点	$Adj.R^2$	*RMSE*/%	数据点
C_{hc} (%, *daf*)	0.9870	1.56	75	0.9422	2.00	19
H_{hc} (%, *db*)	0.9650	0.17	75	0.9632	0.23	19
N_{hc} (%, *db*)	0.9327	0.47	75	0.9323	0.40	19
S_{hc} (%, *db*)	0.9857	0.13	75	0.9254	0.10	19
O_{hc} (%, *db*)	0.9694	1.55	75	0.9038	1.56	19
VM_{hc} (%, *db*)	0.9799	1.94	75	0.9568	3.12	19

Continued

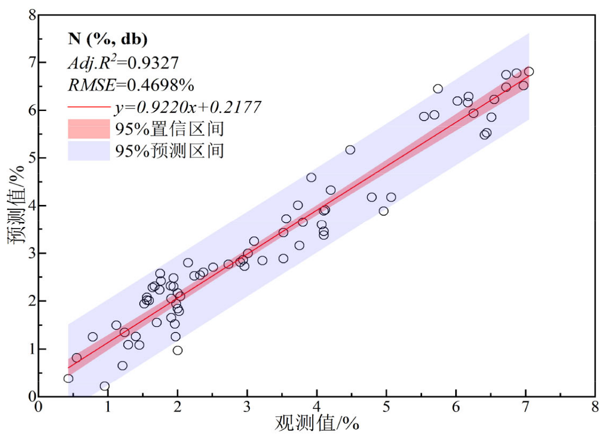
Ash_{hc} (%, *db*)	0.9840	1.51	75	0.9240	3.19	19
FC_{hc} (%, *db*)	0.9074	0.99	75	0.8487	1.11	19
HHV_{hc} (MJ/kg, *db*)	0.8710	1.18	75	0.9263	0.64	19
$Yield_{hc}$ (%)	0.8011	3.16	60	0.8961	4.56	15
MC_{hc} (%)	0.9240	3.71	38	0.7943	6.48	8



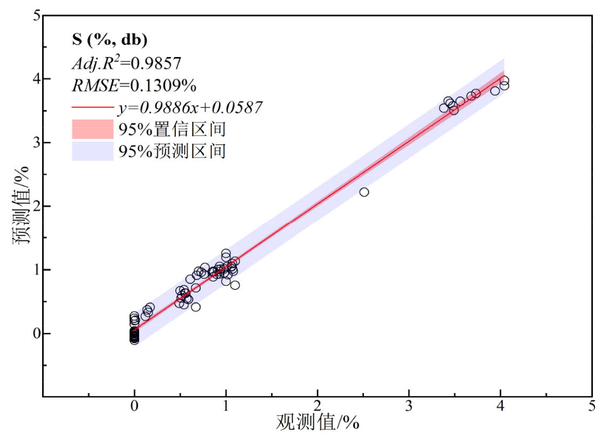
(a)



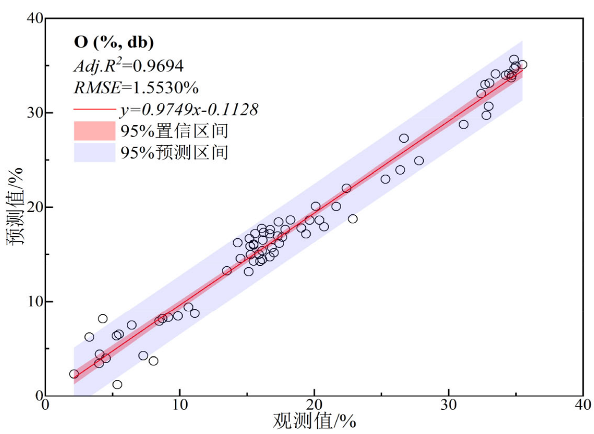
(b)



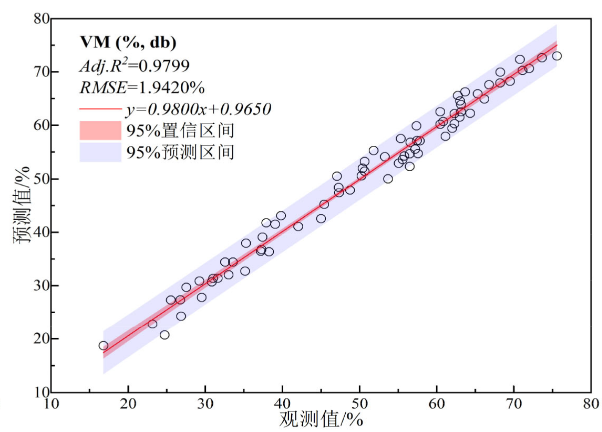
(c)



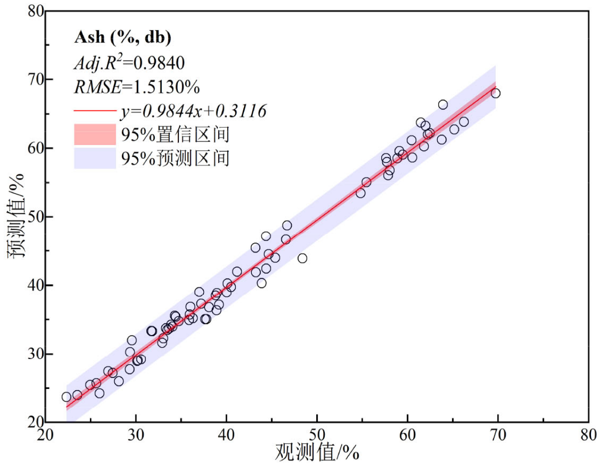
(d)



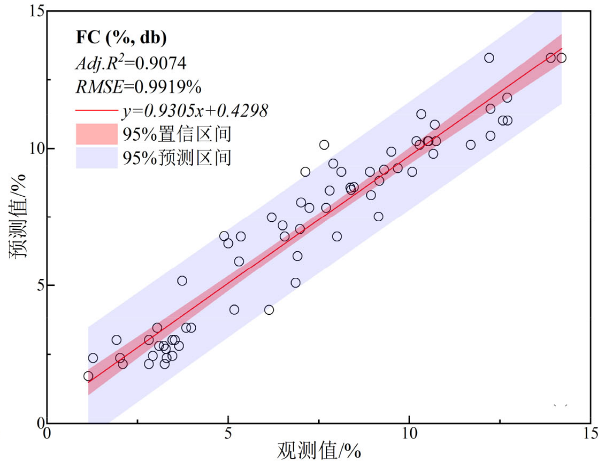
(e)



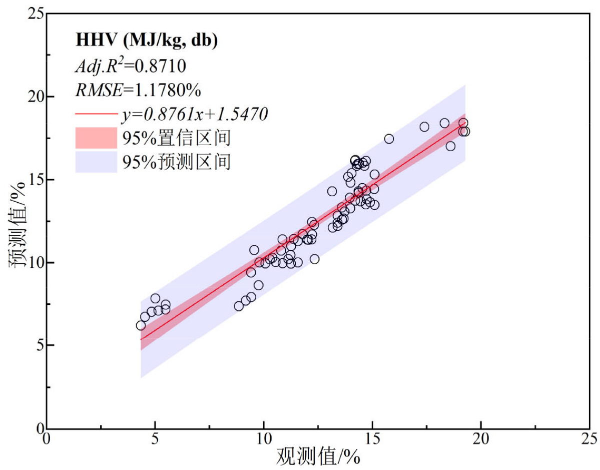
(f)



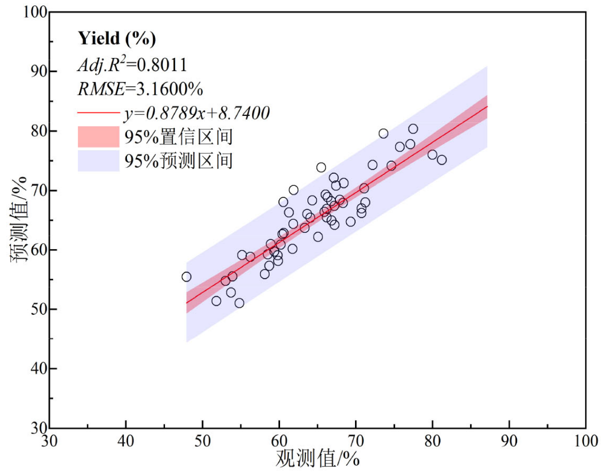
(g)



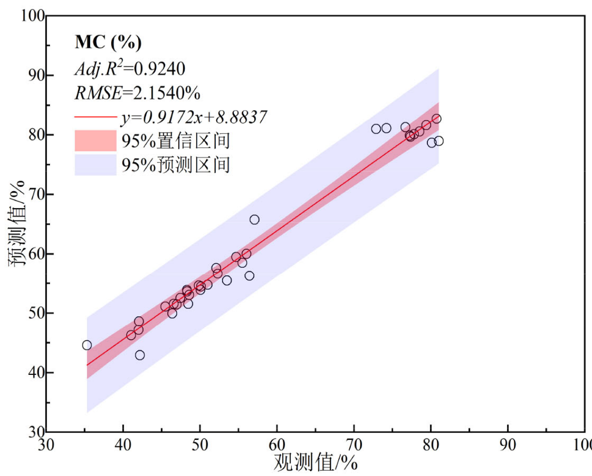
(h)



(i)



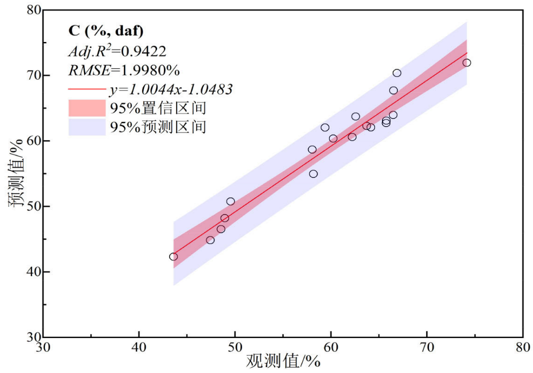
(j)



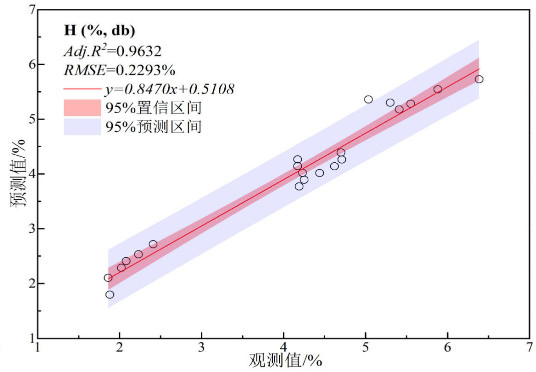
(k)

Figure 4. Comparison of predicted results with experimental data for physiochemical characteristics of hydrochar in the training datasets: (a) C_{hc}, (b) H_{hc}, (c) N_{hc}, (d) S_{hc}, (e) O_{hc}, (f) VM_{hc}, (g) Ash_{hc}, (h) FC_{hc}, (i) HHV_{hc}, (j) $Yield_{hc}$, (k) MC_{hc}

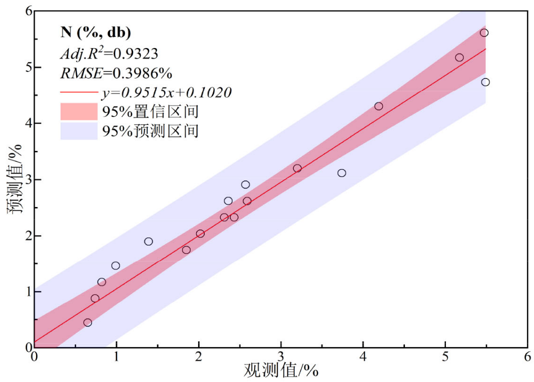
图 4. 训练数据集中水热炭理化性质预测值与实验值比较(a~k 分别为水热炭碳含量、氢含量、氮含量、硫含量、氧含量、挥发分产率、灰分含量、固定碳含量、高位热值、收率和含水率)


C (%, daf)
Adj.R²=0.9422
RMSE=1.9980%
y=1.0044x-1.0483
95%置信区间
95%预测区间
预测值/%
观测值/%

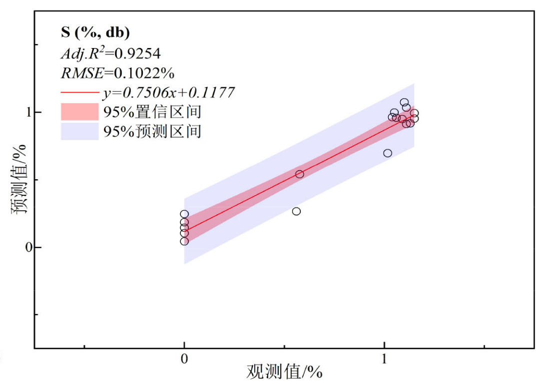
(a)


H (%, db)
Adj.R²=0.9632
RMSE=0.2293%
y=0.8470x+0.5108
95%置信区间
95%预测区间
预测值/%
观测值/%

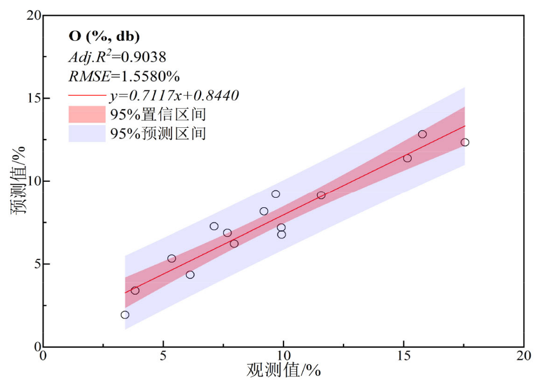
(b)


N (%, db)
Adj.R²=0.9323
RMSE=0.3986%
y=0.9515x+0.1020
95%置信区间
95%预测区间
预测值/%
观测值/%

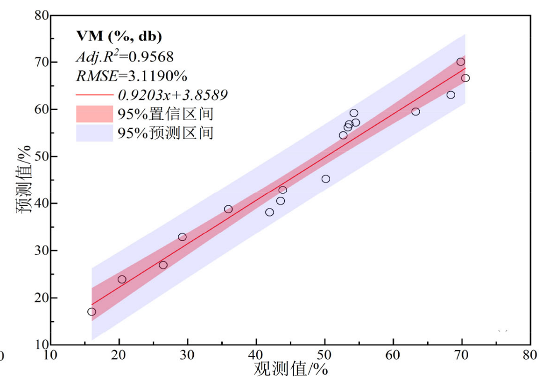
(c)


S (%, db)
Adj.R²=0.9254
RMSE=0.1022%
y=0.7506x+0.1177
95%置信区间
95%预测区间
预测值/%
观测值/%

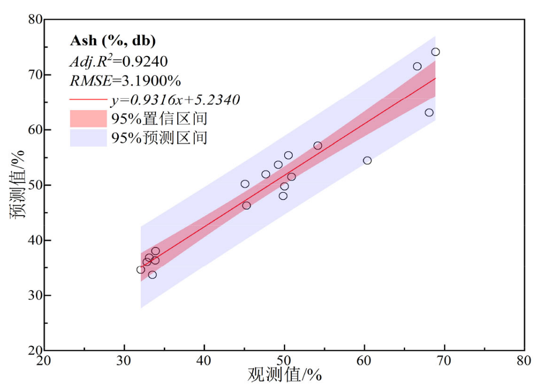
(d)


O (%, db)
Adj.R²=0.9038
RMSE=1.5580%
y=0.7117x+0.8440
95%置信区间
95%预测区间
预测值/%
观测值/%

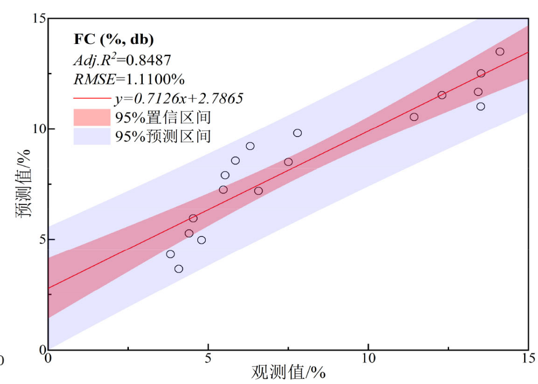
(e)


VM (%, db)
Adj.R²=0.9568
RMSE=3.1190%
0.9203x+3.8589
95%置信区间
95%预测区间
预测值/%
观测值/%

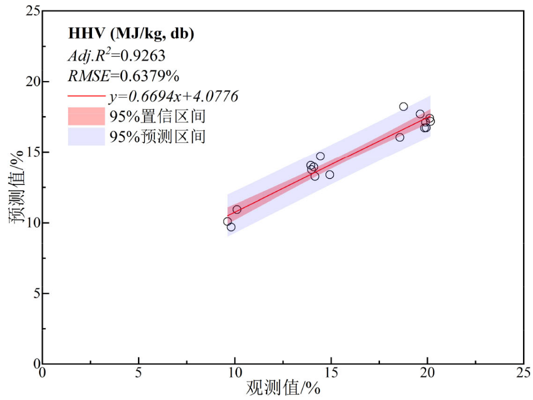
(f)


Ash (%, db)
Adj.R²=0.9240
RMSE=3.1900%
y=0.9316x+5.2340
95%置信区间
95%预测区间
预测值/%
观测值/%

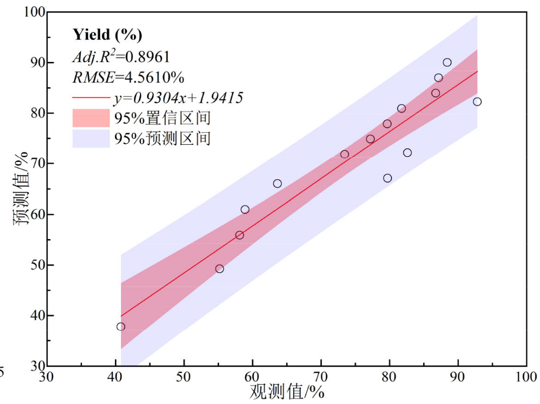
(g)


FC (%, db)
Adj.R²=0.8487
RMSE=1.1100%
y=0.7126x+2.7865
95%置信区间
95%预测区间
预测值/%
观测值/%

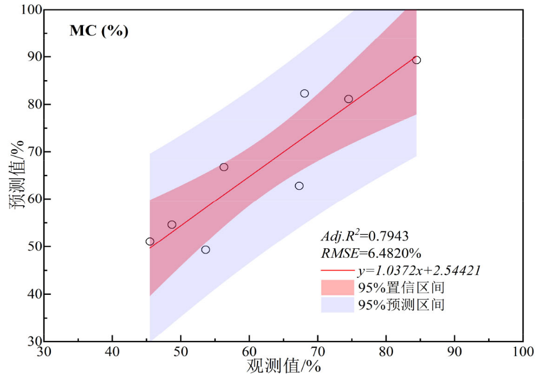
(h)



(i)



(j)



(k)

Figure 5. Comparison of predicted results with experimental data for physiochemical characteristics of hydrocharin the test datasets: (a) C_{hc}, (b) H_{hc}, (c) N_{hc}, (d) S_{hc}, (e) O_{hc}, (f) VM_{hc}, (g) Ash_{hc}, (h) FC_{hc}, (i) HHV_{hc}, (j) $Yield_{hc}$, (k) MC_{hc}

图 5. 测试数据集中水热炭理化性质预测值与实验值比较(a~k 分别为水热炭碳含量、氢含量、氮含量、硫含量、氧含量、挥发分产率、灰分含量、固定碳含量、高位热值、收率和含水率)

4. 结论

1) 皮尔逊相关性分析表明，水热炭中氢、氮、氧和挥发分含量与水热温度具有较强的负相关关系，灰分和固定碳含量与水热温度具有较强的正相关关系。氢含量和氧含量降低是水热处理过程脱水和脱羧基的结果。氮含量降低是因为原始污泥中无机氮和蛋白质发生水解反应和脱氨反应最终形成铵根离子溶解于水热液中。水热处理是一个脱挥发分过程，挥发分产率显著降低，矿物质在水热处理过程富集，灰分含量因而相应地提高。而高温促进聚合反应会导致固定碳含量提高。此外，作为惰性物质的灰分不仅会影响其他物质的变化，还会严重影响水热炭高位热值。剔除高灰分含量的影响，发现水热温度能够促进水热炭中碳含量增加，但较高的水热温度不利于水热炭收率增加。相较于水热温度，反应时间的影响较弱，上述结论与相关实验结果高度一致。

2) 基于方差分析结果的回归模型整体预测精度较高，验证情况较好，除水热炭含水率之外，其他因变量 $Adj.R^2 > 0.8$，$RMSE < 5\%$。建立的水热炭理化性质和收率预测模型可为水热炭燃料特性评估和水热处理条件优化提供指导。

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