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Research Article

Keywords: spontaneous combustion of coal gangue, moisture content, temperature forecasting, particle swarm optimization, gated recurrent unit

Posted Date: October 14th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10805086/v1>

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Additional Declarations: No competing interests reported.

Thermal behavior and oxidation kinetics of coal gangue under varying moisture conditions: An experimental and predictive modeling study

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Abstract

To elucidate the effect of moisture on coal gangue oxidation and spontaneous combustion propensity, samples with five moisture levels (2, 6, 8.61, 12, and 18%) were examined via programmed heating and gas analysis. Multi-dimensional gas indicators (O_2 , CO_2 , $CO/\Delta O_2$, and C_2H_4/C_2H_6) were employed to characterize the oxidation reaction progression and heat release evolution. A hybrid particle swarm optimization–gated recurrent unit (HPSO-GRU) model was developed for self-heating temperature prediction, incorporating moisture content and key gas indicators identified via Spearman correlation as inputs. The predictive performance of HPSO-GRU was comprehensively evaluated against GRU, BO-GRU, IGWO-GRU, PSO-GRU, and SSA-GRU models using a comprehensive performance index (CPI). Thermal analysis revealed that at 8.61% moisture, oxygen consumption and CO generation peaked with maximum heat release, while 18% moisture significantly suppressed exothermic reactions, demonstrating a dual regulatory mechanism of moisture on oxidative self-heating. Molecular dynamics simulations further elucidated that moderate moisture enhances O_2 accumulation around active sites and shortens its diffusion distance,

thereby promoting oxidative exothermy, whereas excessive moisture impedes O₂ mass transfer and occupies active sites, inhibiting heat release. The HPSO-GRU model achieved superior performance (maximum relative error: 7.8%; CPI: 0.0464), substantially outperforming all benchmarks. Feature importance analysis identified O₂ (31.6%) and CO/ Δ O₂ (27.8%) as the dominant predictors, highly consistent with Spearman correlation. A graphical user interface was developed based on the proposed model for rapid internal temperature prediction and intelligent oxidation stage identification of coal gangue piles. This study provides a theoretical foundation and technical support for thermodynamic kinetic assessment and early warning of spontaneous combustion risks in coal gangue.

Keywords:spontaneous combustion of coal gangue; moisture content; temperature forecasting; particle swarm optimization; gated recurrent unit

1 Introduction

Coal gangue, as the primary solid waste generated during coal mining and beneficiation processes, has experienced a continuous increase in accumulation with the sustained development of the coal industry in China[1]. Long-term open-air storage of coal gangue piles promotes progressive heat accumulation within the pile through oxidative reactions, thereby increasing the risk of spontaneous combustion. Coal gangue spontaneous combustion not only releases substantial quantities of toxic and hazardous gases, resulting in severe environmental pollution, but also triggers secondary disasters such as slope instability and explosions, posing significant threats to the safety of surrounding communities and infrastructure[2-3]. Among the various factors governing spontaneous combustion in coal gangue piles, internal temperature represents a critical controlling parameter[4]. Therefore, achieving accurate prediction of internal temperature evolution is of paramount importance for the early warning, prevention, and mitigation of spontaneous combustion hazards in

coal gangue piles.

In recent years, substantial progress has been achieved in the prediction of coal gangue spontaneous combustion temperature. Zheng et al.[5] investigated the spontaneous combustion characteristics of coal gangue under different water immersion ratios through programmed temperature oxidation experiments and gas analysis. Deng et al.[6] systematically analyzed the combustion characteristics during the oxidative heating process of coal gangue and identified seven characteristic temperature points and four distinct combustion stages. Zhou et al.[7] further elucidated the evolution mechanisms of oxidation-sensitive functional groups in coal gangue under varying water immersion conditions through experimental investigations. Meanwhile, research methodologies developed for coal spontaneous combustion have provided valuable insights into understanding the oxidation and combustion mechanisms of coal gangue. Deng et al.[8] conducted programmed heating experiments on different coal samples and employed mathematical approaches to investigate the relationships between indicator gases and characteristic temperatures. Guo et al.[9] monitored gas concentrations during combustion and demonstrated the feasibility of gas composition analysis for temperature prediction. Experimental results reported by Beamish et al.[10] revealed that when moisture content exceeds the critical water-holding capacity, the oxidation process is almost completely inhibited. Shi et al.[11] further confirmed that low moisture levels enhance the reaction kinetics between oxygen and reactive functional groups, whereas excessive moisture tends to block pore channels and occupy active sites, thereby impeding oxygen adsorption and diffusion. These studies collectively demonstrate that moisture content exerts a significant influence on the oxidative heat release characteristics of coal and coal gangue. However, the inherent nonlinear correlations between indicator gases and spontaneous combustion temperature remain a major challenge, limiting prediction accuracy. Therefore, elucidating the complex nonlinear mechanisms governing gas

evolution and temperature variation is essential for improving the reliability and precision of spontaneous combustion temperature prediction.

Machine learning and advanced computational algorithms have increasingly been employed to elucidate the complex nonlinear relationships between gas concentration evolution and combustion processes[12-14]. In recent years, deep learning frameworks have evolved from early single-model architectures toward more sophisticated and integrated hybrid structures. Deng et al.[15] employed grid search techniques combined with a support vector machine (SVM) classification algorithm to optimize model parameters and developed a predictive model applicable to multiple coal types. However, the performance of SVM models remains highly sensitive to parameter selection. Yan et al.[16] proposed an improved radial basis function(RBF) neural network for spontaneous combustion prediction and early warning in goaf areas. Although this approach accelerated network convergence, it remained susceptible to overfitting. Chen et al.[17] utilized an improved grey wolf optimizer(IGWO) to optimize and compare several models, including back propagation neural networks(BPNN), convolutional neural networks(CNN), and gated recurrent units(GRU), demonstrating that the GRU model achieved the best overall performance. Benefiting from update and reset gate mechanisms, GRU exhibits superior capability in sequential data prediction. However, its predictive accuracy strongly depends on hyperparameter configuration, manual tuning, and grid-search strategies, resulting in low computational efficiency and limited generalization capability. Although the incorporation of swarm intelligence optimization algorithms enables adaptive parameter optimization to some extent, these methods remain vulnerable to local optima. The hybrid particle swarm optimization (HPSO) algorithm integrates multiple enhancement strategies, including chaotic initialization, adaptive inertia weighting, gradient descent, local search, elite preservation, and periodic restart mechanisms. These improvements effectively overcome the premature convergence

and population diversity degradation issues associated with conventional PSO, achieving a dynamic balance between global exploration and local exploitation capabilities[18].

Although considerable progress has been achieved in spontaneous combustion temperature prediction, existing studies still face several challenges associated with data acquisition, model generalization capability, and prediction accuracy. Moreover, moisture content plays a critical role in regulating the oxidation behavior of coal gangue; however, systematic investigations into the spontaneous combustion propensity of coal gangue under different moisture conditions remain insufficient. Therefore, this study systematically investigates the oxidation behavior of coal gangue from three perspectives: macroscopic oxidation characteristics, microscopic oxygen diffusion mechanisms, and intelligent prediction modeling. The nonlinear evolution patterns of coal gangue oxidation reactivity under varying moisture conditions are elucidated, and the microscopic mechanisms by which moisture regulates oxygen diffusion and reactions at active sites are revealed. Furthermore, an HPSO-GRU-based temperature prediction model for coal gangue is developed, providing theoretical insights and technical support for early warning and risk mitigation of spontaneous combustion hazards in coal gangue piles.

2 Experimental Scheme and Simulation Methodology

2.1 Programmed Temperature Rise Experiment

The coal gangue samples used in this study were collected from the mining and excavation working faces of the Shendong mining area. Fresh coal gangue samples were selected, and the surface oxidation layer was removed prior to subsequent experiments. The samples were first crushed using a jaw crusher and then ground to a particle size range of 70-110 mesh. Subsequently, the samples were dried in a forced-air drying oven at 105°C until a constant weight was achieved, obtaining a

completely dried state. A portion of the dried samples was subjected to water immersion treatment. When the samples reached water saturation, and their mass remained constant, the saturated moisture content was determined to be 19.21%. Based on the dry samples, distilled water was gradually and uniformly sprayed onto the samples through an intermittent mixing process to prepare specimens with target moisture contents of 2%, 6%, 8.61% (original samples without water immersion), 12%, and 18%. The moisture content was calculated according to Equation (1)[19]:

$$W = \frac{m_w}{m_w + m_d} \times 100\% \quad (1)$$

where W represents the moisture content of the sample, m_w denotes the mass of distilled water required, and m_d corresponds to the absolute dry mass of the sample.

A total of 80 g of each prepared coal gangue sample with different moisture contents was weighed and placed into a coal pyrolysis-gasification integrated experimental apparatus (Fig.1). The YKRL electric heating furnace was initially maintained at 30 °C and heated to 550°C at a heating rate of 1°C/min, with a stabilization period of 3 min at each target temperature. During heating, dry air was continuously introduced at a flow rate of 100 mL/min after each 1°C increment, and the released gases were collected in gas sampling bags. Subsequently, the gas compositions were analyzed using a GC-4000A gas chromatograph.

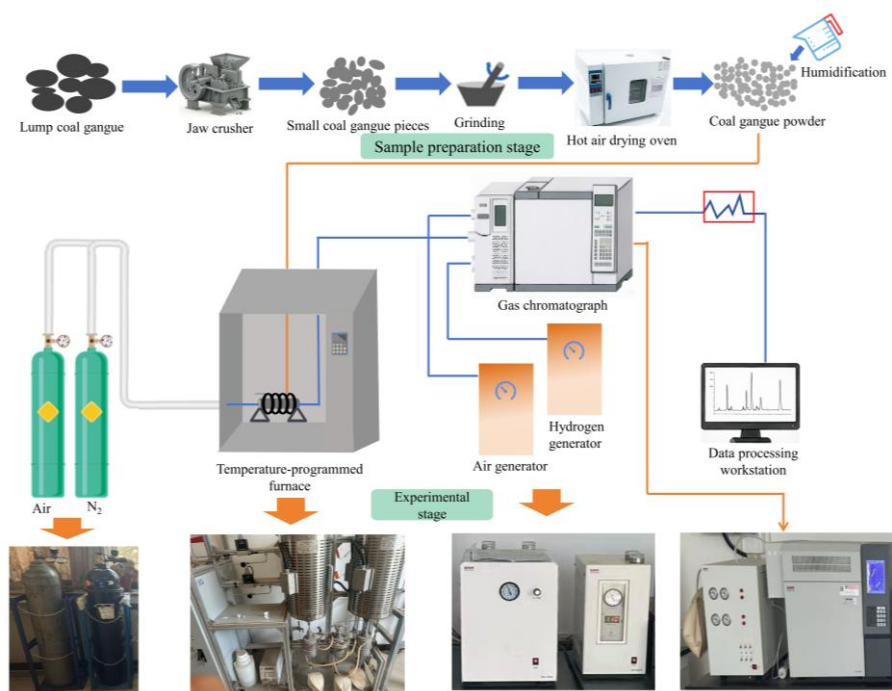


Fig. 1 Schematic diagram of the temperature-programmed experiment

2.2 Molecular Dynamics(MD) Simulation of Coal Gangue

The primary mineral components of coal gangue mainly include kaolinite and quartz, among which kaolinite is one of the most abundant and structurally representative mineral phases. Previous studies have demonstrated that the layered structure of kaolinite contains abundant surface-active sites, such as hydroxyl groups(-OH), which can interact with oxygen and water molecules via adsorption and exert a significant influence on oxidation reactions.

2.2.1 Model Selection and Construction

To elucidate the microscopic regulatory mechanism of moisture content on coal gangue oxidation reactivity at the molecular scale, kaolinite was selected as a simplified mineral framework model representing the primary mineral phase of coal gangue. The three-dimensional molecular structures were constructed using the Amorphous Cell module in Materials Studio(MS). Under three-dimensional periodic boundary conditions, the kaolinite mineral framework and varying amounts of H₂O molecules were introduced into cubic simulation cells to generate coal gangue-water composite

systems. By adjusting the number of incorporated water molecules, molecular models corresponding to moisture contents of 2%, 8.61%, and 18% were accurately established. The initial density of the simulation cells was set to 1.20 g/cm³, with dimensions of 2 nm×2 nm×1 nm.

2.2.2 Simulation Calculation

Molecular dynamics simulations were performed using the COMPASS II all-atom force field. The initial configurations were first subjected to energy minimization using the Smart Minimizer algorithm to eliminate local steric hindrance and obtain energetically stable structures. Subsequently, simulations were conducted under the NVT ensemble for 500 ps, with temperature and pressure regulation achieved using the Nose-Hoover thermostat and Andersen method, respectively. After structural equilibration, an additional 5000 ps simulation was carried out to collect trajectory data, with a time step of 1.0 fs. The simulation temperatures were set at 120, 310, and 460°C, corresponding to three typical characteristic temperatures identified in the programmed temperature rise experiments. Electrostatic interactions and van der Waals interactions were calculated using the Ewald summation method, with a cutoff radius of 1.25 nm for both interactions.

2.2.3 Parameter Calculation

Based on the dynamic trajectories obtained from the simulations, key kinetic and structural parameters were calculated by statistically averaging the positions and velocity vectors of all particles within the simulation cells. The following critical parameters were subsequently determined[20-21] :

(1) Radial Distribution Function(RDF)

The $g(r)$ was employed to quantitatively characterize the localized heterogeneous aggregation behavior and spatial arrangement of H₂O molecules on the surfaces of active functional groups in the coal gangue molecular system. The calculation equation is expressed as follows (Eq. 2):

$$g(r) = \frac{1}{4\pi\rho_B r^2} \frac{dN}{dr} \quad (2)$$

where r represents the distance between particle A and particle B, nm; ρ_B denotes the number density of particle B, nm; and dN is the average number of particle B located within the radial distance range from r to $r+dr$ around particle A, nm.

(2) Mean Square Displacement (MSD) and Diffusion Coefficient (D_c)

MSD and D_c were employed to quantitatively evaluate the microscopic diffusion dynamics of H₂O molecules under different moisture contents and temperature conditions. The calculation equation is given as follows (Eq. 3):

$$MSD = \frac{1}{N} \sum_{i=1}^N [r_i(t) - r_i(0)]^2 \quad (3)$$

$$D_c = \frac{1}{6N} \lim_{t \rightarrow \infty} \frac{d}{dt} \sum_{i=1}^N [r_i(t) - r_i(0)]^2 \quad (4)$$

where N represents the total number of diffusing molecules, and $r(t)$ denotes the position vector of a molecule at time t .

3 HPSO-GRU Prediction Model

During the oxidative heating of coal gangue, gas indicators exhibit significant stage-dependent and time-sequential responses to temperature variations. The prediction performance of conventional GRU models is highly sensitive to hyperparameter configurations, limiting their adaptability under complex operating conditions with varying moisture contents. Therefore, the HPSO algorithm was introduced to achieve global adaptive optimization of key GRU hyperparameters, thereby improving the prediction accuracy and generalization capability of the model.

3.1 Hybrid Particle Swarm Optimization (HPSO)

The HPSO algorithm integrates multiple strategies, including parameter adaptation, chaotic

initialization, gradient optimization, local search, quantum perturbation, and elite preservation, to establish a multi-module collaborative optimization framework. The chaotic mechanism enhances population diversity, the elite strategy improves convergence efficiency, the gradient optimization strategy increases local search accuracy, and the periodic restart mechanism prevents premature convergence[22]. Compared with conventional PSO, HPSO effectively improves search stability and global optimization capability in complex optimization tasks, achieving a dynamic balance between exploration and exploitation[23-24]. The update strategy of the inertia weight is expressed as follows:

$$w(t) = w_{\max} - (w_{\max} - w_{\min}) \cdot \left(\frac{t}{T_{\max}}\right)^{0.5} \quad (5)$$

The individual cognitive coefficient and social coefficient are determined by Eqs. (6) and (7), respectively:

$$c_1 = c_{1,i} - (c_{1,i} - c_{1,f}) \frac{t}{T} \quad (6)$$

$$c_2 = c_{2,i} + (c_{2,i} - c_{2,f}) \frac{t}{T} \quad (7)$$

where $c_{1,i}$ and $c_{1,f}$ represent the initial and final individual cognitive coefficients, respectively, while $c_{2,i}$ and $c_{2,f}$ denote the initial and final social coefficients, respectively.

The velocity update strategy is determined by Eq. (8) as follows:

$$v_i^{t+1} = w(t)v_i^t + c_1(t)r_1(pbest_i - x_i^t) + c_2(t)r_2(gbest - x_i^t) \quad (8)$$

The position update strategy is determined by Eq. (9) as follows:

$$x_i^{t+1} = x_i^t + wv_i^t + c_1r_1(p_i - x_i^t) + c_2r_2(g - x_i^t) - \eta \nabla f(x_i^t) + \alpha_q(rand - 0.5)(ub - lb) \quad (9)$$

where $f(x)$ represents the fitness function; $\nabla f(x_i^t)$ denotes the gradient of the objective function; η is the gradient learning rate; and $(rand-0.5)$ is a random perturbation term within the range of $[-0.5, 0.5]$.

3.2 Gated Recurrent Unit (GRU) Algorithm

As a lightweight variant of recurrent neural networks (RNNs), the GRU model effectively overcomes the gradient vanishing and exploding problems of conventional RNNs by introducing update and reset gates, enabling the modeling of long-term temporal dependencies[25]. Compared with LSTM, GRU has a simpler architecture, fewer parameters, and higher training efficiency, while maintaining strong prediction performance and reducing the risk of overfitting. Through its gating mechanism, GRU dynamically regulates the proportion of historical information retained and optimizes hidden state updates. The calculation process is expressed as Eq. (10):

$$\begin{cases} z_t = \sigma(W_z \cdot [h_{t-1}, x_t]) \\ r_t = \sigma(W_r \cdot [h_{t-1}, x_t]) \\ h_t = \tanh(W \cdot [r_t * h_{t-1}, x_t]) \\ h_t = h_{t-1} * (1 - z_t) + h_t * z_t \end{cases} \quad (10)$$

where σ denotes the Sigmoid activation function; z_t and r_t represent the update gate and reset gate, respectively; W_z , W_r and W are the corresponding neuron weight parameters of each gate; H_t represents the hidden state of the memory unit; Tanh denotes the hyperbolic tangent activation function; and X_t represents the input feature vector at the current time step.

3.3 Development of the HPSO-GRU Temperature Prediction Model

To reveal the intrinsic relationship between the temporal variations of gas indicators and the temperature evolution of coal gangue, an HPSO-GRU temperature prediction model was developed. The proposed model uses multi-dimensional gas indicators as input features and temperature as the prediction target. The GRU network is employed to extract nonlinear temporal characteristics between gas evolution and temperature changes, while the HPSO algorithm adaptively optimizes key hyperparameters, such as the number of hidden neurons and learning rate. The minimum RMSE obtained from five-fold cross-validation is adopted as the optimization objective to achieve adaptive

adjustment of the model structure. The proposed model consists of several key procedures, including data preprocessing, GRU-based feature extraction, HPSO-driven hyperparameter optimization, and performance validation (Fig. 2). The optimized model was further compared with GRU models coupled with different optimization algorithms to comprehensively evaluate its prediction accuracy and robustness.

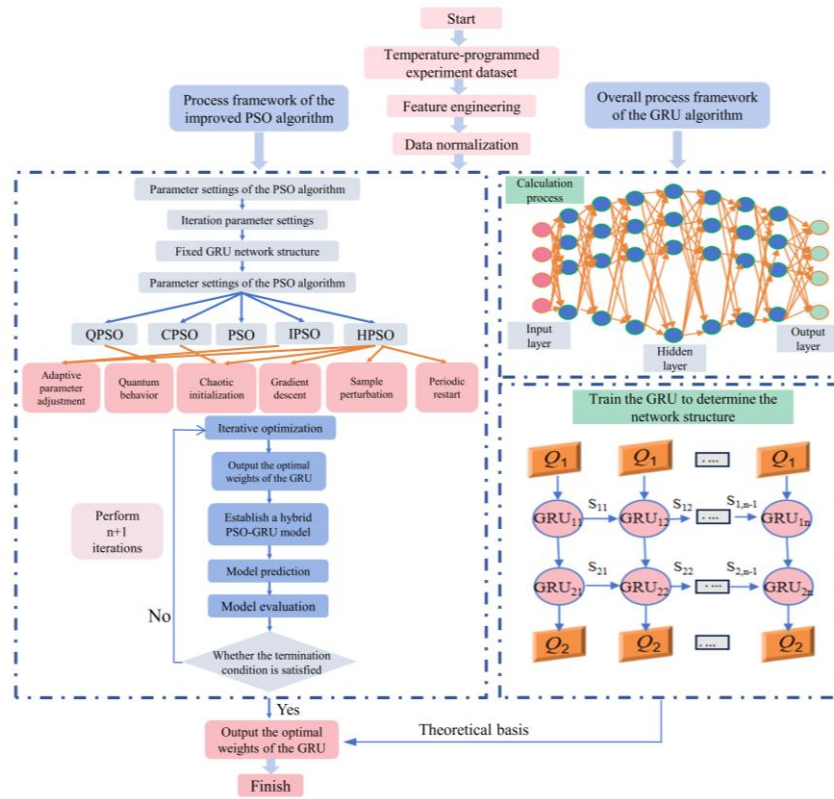


Fig. 2 Principles and prediction flow of the HPSO-GRU model

3.4 Model Training

The program-heating experimental data were automatically processed using Python, including the identification and removal of duplicate records, abnormal values, and missing samples. After rigorous data screening, a total of 2254 valid data points were obtained (Table 1). The dataset was randomly divided into training and testing sets at a ratio of 7:3 for model development and performance evaluation. The gas indicators and moisture content were selected as input features, while temperature was used as the prediction target. To eliminate the dimensional differences among

input variables and accelerate model convergence, the MinMaxScaler method (Eq. 11) was applied to normalize all input features[26].

$$X_s = \frac{X - X_{\min}}{X_{\max} - X_{\min}} \quad (11)$$

where X_s represents the normalized value; $X_{\max}$ and $X_{\min}$ denote the maximum and minimum values of variable X , respectively.

Table1 Brief description of the training dataset

Temperature/°C	Moisture Content/%	O ₂ /%	CO/ppm	CO ₂ /ppm	C ₂ H ₄ /ppm	CO/ΔO ₂	C ₂ H ₄ /C ₂ H ₆
30	2%	20.52	0	517	0	0	0
31	2%	20.51	0	519	0	0	0
...							
152	2%	20.12	43.42	741	0.01159	0.722	0.643
...							
549	18%	16.21	4 796	10 826	0.3244	3.214	1.861
550	18%	16.21	4 813	10 911	0.3219	3.243	1.855

4 Results Analysis and Model Performance Validation

4.1 Gas Evolution Characteristics of Coal Gangue with Different Moisture Contents

4.1.1 Stage-Specific Characteristics of Individual Gas Products

Figure 3 illustrates the variation patterns of the volume fractions of O₂, CO₂, CO, CH₄, C₂H₆, and C₂H₄ during the heating process. With increasing temperature, the O₂ concentration continuously decreases due to gas desorption, decomposition, and consumption reactions. At approximately 120°C, the decline rate of O₂ concentration increases sharply, indicating the transition of coal gangue into the rapid oxidation stage. When the temperature exceeds 140°C, the differences in outlet oxygen concentrations among samples with different moisture contents become increasingly pronounced, with the oxygen consumption rate following the order: $W = 8.61\% > 6\% > 12\% > 2\% > 18\%$. As the temperature rises to approximately 460°C, the combustible components in coal gangue, including

volatile matter and fixed carbon, are almost completely consumed, marking the transition into the burnout stage, during which the oxygen consumption rate gradually decreases. These results demonstrate that moisture content exerts a dual regulatory effect on the oxidation process of coal gangue: moderate moisture levels promote oxidation by enhancing oxygen consumption, whereas excessive moisture suppresses oxidation reactions.

The concentrations of oxygen-containing carbonaceous compounds and hydrocarbon gases exhibit a continuous increasing trend with rising temperature. Among these gases, CO and CO₂ are first detected at 95°C and 30°C, respectively. C₂H₆ begins to appear at approximately 120°C, primarily originating from the thermal cracking of aliphatic side chains within coal gangue, and can be regarded as an indicator of the initial pyrolysis stage. During this period, the gas yield remains relatively low, indicating that oxidation reactions have not yet developed extensively.

When the temperature increases to approximately 150°C, C₂H₄ is generated through the high-temperature cracking of long-chain hydrocarbons and serves as an important indicator gas during the oxidative heating process, suggesting that coal gangue has entered the rapid oxidation stage. Therefore, before 120°C, coal gangue mainly undergoes a slow oxidation process. However, after 310°C, the concentrations of CO, CO₂, C₂H₄, and C₂H₆ increase dramatically, indicating that coal gangue has transitioned into an intensive oxidation stage.

During the slow oxidation stage, the CO and CO₂ production rates of coal gangue samples with different moisture contents remain relatively low, and the differences among samples are insignificant. However, upon entering the rapid oxidation stage, the variations in CO and CO₂ generation become increasingly pronounced. Among all samples, the specimen with a moisture content of $W=8.61\%$ exhibits the highest CO production rate, whereas the $W=18\%$ sample shows the lowest value. Consistent with the generation characteristics of carbon oxides, an appropriate moisture level

promotes the decomposition of reactive functional groups and facilitates the formation of hydrocarbon products at elevated temperatures. In contrast, excessive moisture can cover surface active sites, hinder oxygen accessibility, and suppress deep oxidation reactions, thereby reducing the release of hydrocarbon gases.

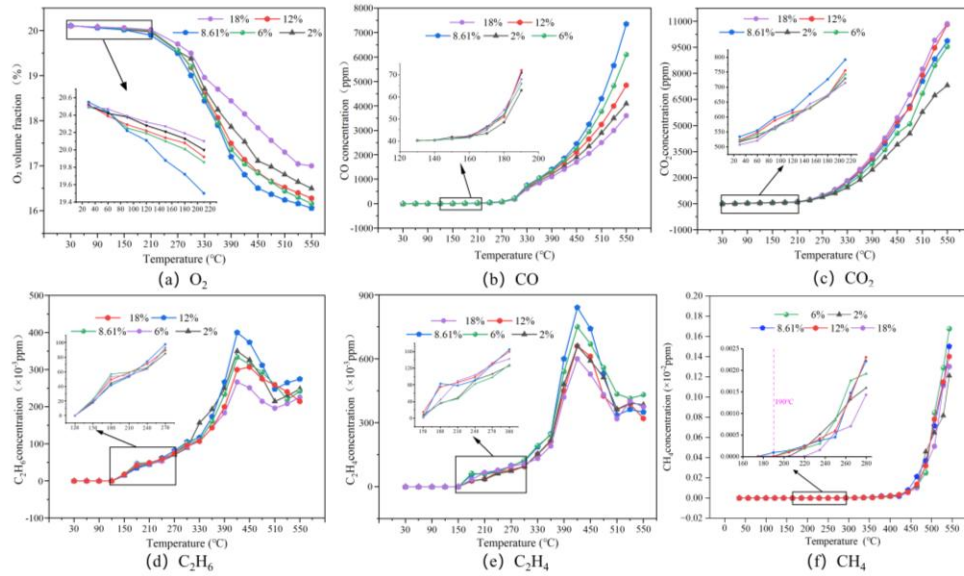


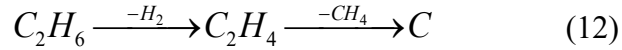
Fig. 3 Variation of individual gas concentrations with temperature

4.1.2 Evolution Patterns of Composite Gas Characteristic Ratios

In coal gangue piles, individual gas concentrations may be affected by internal air circulation, resulting in prediction deviations. Therefore, composite gas parameters were introduced as auxiliary indicators to improve prediction accuracy. Due to the relatively low concentrations of C_2H_4 , C_2H_6 , and CH_4 , and the relatively stable variation of ΔO_2 compared with CO generation during the heating process, four composite gas indices, namely C_2H_4/C_2H_6 , $CO/\Delta O_2$, CH_4/C_2H_6 , and CO_2/CO , were incorporated into the gas-based prediction framework from the perspective of oxidation reaction intensity.

As shown in Fig. 4(c), the C_2H_4/C_2H_6 ratio exhibits an initial increase followed by a subsequent decline with increasing temperature. At approximately 220°C, the direct cracking reaction during the

secondary oxidation process of coal gangue promotes the conversion of C_2H_6 into C_2H_4 (Eq. 12), resulting in a rapid increase in the ratio. When the temperature exceeds 440°C , C_2H_4 undergoes further transformation into C_2H_6 , leading to a decrease in the C_2H_4/C_2H_6 ratio.



The magnitude of $CO/\Delta O_2$ is commonly used as an indicator of oxidation intensity. As illustrated in Fig. 4(b), $CO/\Delta O_2$ exhibits an overall increasing trend with increasing temperature. Before 120°C , coal gangue remains in a slow low-temperature oxidation stage dominated by weak physical desorption, resulting in relatively minor fluctuations in the ratio. When the temperature exceeds 120°C , accelerated oxidation leads to a significant decrease in O_2 concentration, thereby increasing the $CO/\Delta O_2$ ratio. At approximately 180°C , the decomposition of oxygen-containing functional groups, such as carbonyl ($>C=O$) and quinone groups, generates abundant free radicals, further promoting a rapid increase in $CO/\Delta O_2$. When the temperature reaches approximately 470°C , as described by Eqs. (13)-(15), the decomposition of oxygen-containing functional groups produces a large amount of CO , resulting in another sharp increase in the $CO/\Delta O_2$ ratio.

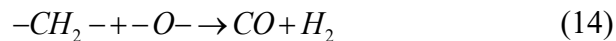
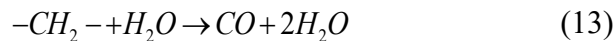


Figure 4(a) shows that the CO_2/CO ratio initially increases and then decreases with increasing temperature during the early stage of the experiment. At approximately 250°C , the ratio gradually stabilizes, followed by a continuous decline, reaching a minimum value around 350°C before increasing again. The CO_2/CO ratio reaches its peak at 150°C , indicating a rapid increase in CO_2 generation, with its production rate significantly exceeding that of CO at this stage. As shown in Fig. 4(d), the C_2H_4/C_2H_6 ratio continuously increases with increasing temperature. After 220°C , the

growth rate of the curve becomes markedly accelerated, which is primarily attributed to the enhanced desorption and release of CH_4 at elevated temperatures. Since the generation rate of CH_4 is substantially higher than that of C_2H_6 , the ratio between the two gases increases rapidly.

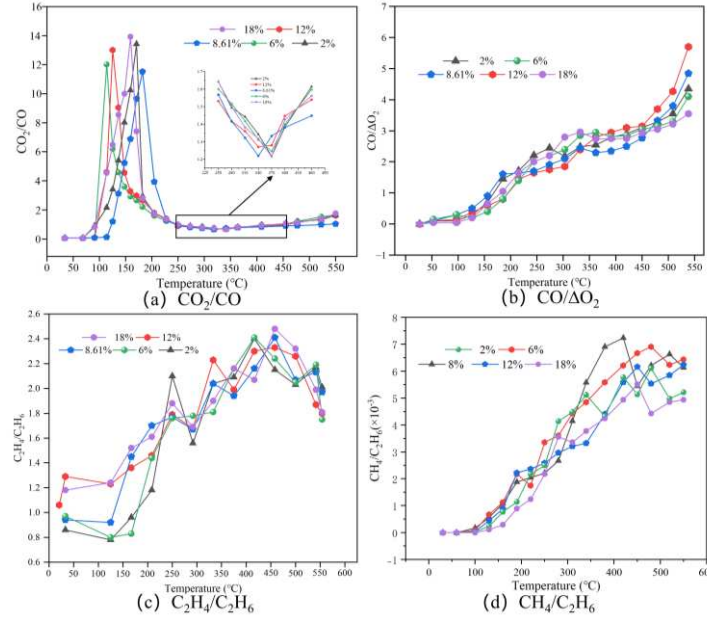


Fig. 4 Variation of composite gas indices with temperature

4.2 MD-Based Analysis of the Moisture Regulation Mechanism

As shown in Fig. 5, the RDF curves of H_2O molecules under different moisture contents and temperature conditions exhibit similar profiles, indicating that they follow comparable adsorption behaviors. However, significant differences are observed in the intensity of the characteristic peaks. At $T=310^\circ\text{C}$ and $W=8.61\%$, the maximum RDF peak intensity between H_2O molecules and active sites of coal gangue reaches 8.01, which is substantially higher than those of the $W=2\%$ (6.40) and $W=18\%$ (4.48) systems. Across all simulated temperatures, the RDF peak intensities follow the same order: $W=8.61\% > W=2\% > W=18\%$. This indicates that H_2O molecules exhibit the strongest localized aggregation around active sites under the $W=8.61\%$ condition. Considering the competitive adsorption relationship between H_2O and O_2 molecules on coal gangue surfaces, a higher degree of H_2O aggregation suggests stronger molecular confinement and enhanced adsorption capacity of the surface region, thereby facilitating the capture and enrichment of O_2 molecules near reactive centers.

The MSD curves demonstrate that, during different oxidation stages, the MSD values initially increase and subsequently decrease with increasing moisture content. The average MSD value reaches a maximum of 5598.47 Å² at $W=8.61\%$, representing increases of 1172.45 Å² and 2350.99 Å² compared with the $W=2\%$ and $W=18\%$ systems, respectively. The variation trend of the D_c is consistent with that of MSD, following the order: $W=8.61\%$ (2.78×10^{-4} m²/s) > $W=2\%$ (2.26×10^{-4} m²/s) > $W=18\%$ (1.62×10^{-4} m²/s). These results demonstrate that H₂O molecules possess the strongest diffusion capability and collision-induced activation effect at $W=8.61\%$, which provides the most favorable conditions for O₂ penetration and migration. In contrast, excessive moisture at $W=18\%$ inhibits the approach of O₂ molecules toward active sites.

The combined analysis of RDF and MSD results reveals that moisture content exerts a dual regulatory effect on the oxidation behavior of coal gangue. An appropriate moisture level can effectively enhance the mobility of surface O₂ molecules, promoting their penetration into the micro-scale pore structures of coal gangue and consequently increasing its spontaneous combustion tendency. Molecular configuration analysis further indicates that at $W=8.61\%$, water molecules act as bridging structures between oxygen-containing functional groups, forming stable cyclic hydrogen-bond networks while maintaining relatively unobstructed pore channels. Conversely, at $W=18\%$, excessive water molecules generate a thick physically adsorbed water layer on the coal gangue surface, significantly narrowing pore pathways and encapsulating part of the active reaction sites. .

In summary, integrating the programmed temperature rise experiments with molecular dynamics simulations demonstrates that variations in moisture content primarily regulate the oxidation process by influencing oxygen molecular transport behavior. The structural evolution of water molecules can effectively reflect changes in O₂ adsorption capacity, thereby quantitatively characterizing the differences in spontaneous combustion tendency of coal gangue under varying moisture conditions.

Accordingly, the moisture content can be classified into three distinct regimes: an oxidation-promoting region ($W < 6\%$), a highly oxidative region ($6\% \leq W \leq 12\%$), and an oxidation-inhibiting region ($W > 12\%$). This classification provides a molecular-scale theoretical basis for the early prediction and risk assessment of spontaneous combustion in coal gangue piles.

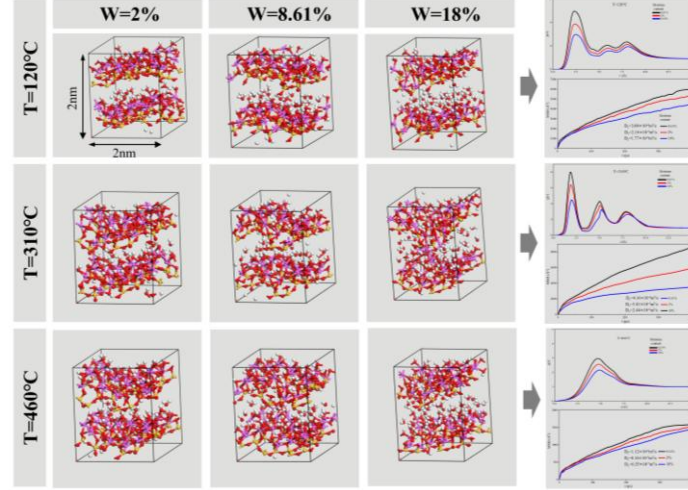


Fig. 5 Molecular mechanism of coal gangue spontaneous combustion under different moisture contents

4.3 Determination of Prediction Indicators

To identify the key gas indicators exhibiting strong correlations with temperature, the Spearman correlation coefficient was employed to evaluate the relationships between gas parameters and temperature under different moisture content conditions[27]. Five coal gangue samples with moisture contents of 2%, 6%, 8.61%, 12%, and 18% were selected. A total of 10 gas-related parameters, including individual indicators (O_2 , CO_2 , CO , CH_4 , C_2H_6 , and C_2H_4) and composite indicators (C_2H_4/C_2H_6 , $CO/\Delta O_2$, CH_4/C_2H_6 , and CO_2/CO), were systematically analyzed to determine their correlation coefficients with temperature. The calculation equation is given as follows (Eq. 16):

$$r_s = 1 - 6 \sum_{i=1}^n d_i^2 / [n(n^2 - 1)] \quad (16)$$

where d_i represents the rank difference between each pair of observations, n denotes the sample size, and the value range of ρ is $[-1, 1]$. A larger absolute value of ρ indicates a stronger monotonic relationship between the variables.

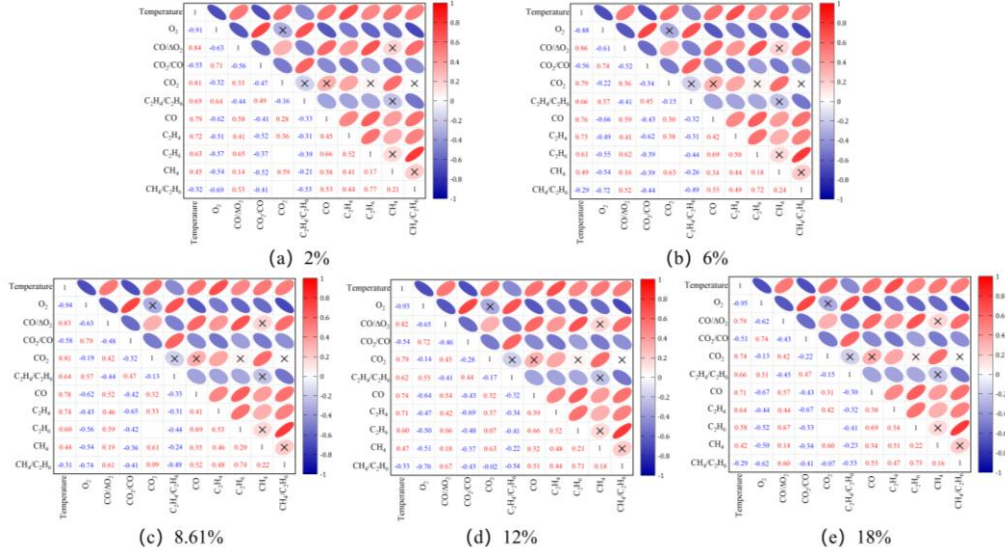


Fig. 6 Correlation between temperature and multi-gas indices for coal gangue at different moisture contents

As shown in Fig. 6, the response characteristics of gas indicators to temperature are generally consistent under different moisture content conditions. The only exception is that the correlation coefficient of C_2H_4/C_2H_6 exceeds that of C_2H_4 when the moisture content reaches 18%. According to the ranking of the absolute correlation coefficients between each indicator and temperature, O_2 , $CO/\Delta O_2$, CO_2 , CO , C_2H_4 , and C_2H_4/C_2H_6 exhibit strong correlations with temperature, whereas CH_4 , CH_4/C_2H_6 , and CO_2/CO show relatively weak correlations. Therefore, these six strongly correlated indicators were selected as the primary gas input features for the subsequent temperature prediction model.

4.4 Prediction Performance Analysis of the HPSO-GRU Model

To quantitatively evaluate the prediction performance of the HPSO-GRU model, the test dataset was input into the proposed model, and the predicted results were compared with those obtained from several benchmark models, including GRU, BO-GRU, IGWO-GRU, PSO-GRU, and SSA-GRU.

Figure 7 presents the prediction results of different models on a subset of the test dataset. As illustrated, the prediction curve generated by the HPSO-GRU model exhibits a high degree of agreement with the actual temperature curve, demonstrating superior fitting performance. In contrast, the SSA-GRU model shows considerable fluctuations during the initial prediction stage, resulting in

significant deviations between predicted and experimental values. Furthermore, when the temperature exceeds 240°C, the prediction errors of the GRU, SSA-GRU, and IGWO-GRU models become increasingly pronounced, thereby reducing prediction accuracy. Compared with these models, the HPSO-GRU model maintains the smallest deviation throughout the entire prediction process and achieves high prediction accuracy for critical temperature points. The maximum relative error of the HPSO-GRU model on the test dataset is only 7.8%, which is significantly lower than that of the five benchmark models. These results demonstrate the robust prediction capability and effective error control performance of the proposed HPSO-GRU model.

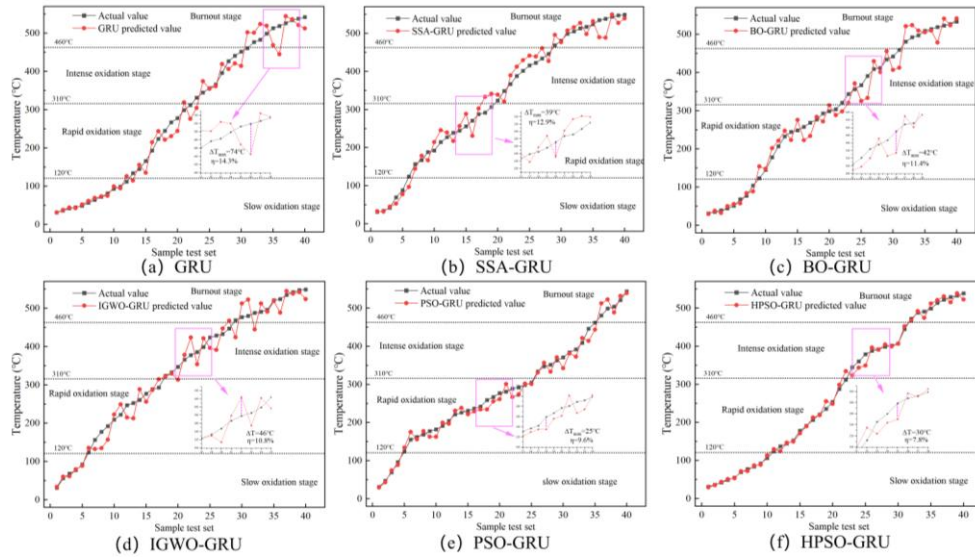


Fig. 7 Comparison of prediction accuracy of different models

4.5 Comprehensive Performance Evaluation of Multiple Prediction Models

To further evaluate the predictive generalization capability of the HPSO-GRU-based forecasting model, the test dataset was applied to the aforementioned six models for comparative analysis. Five classical performance evaluation metrics were employed to comprehensively assess the prediction accuracy and fitting capability of each model from different perspectives[28]. The calculation formulas are as follows:

$$R^2 = 1 - \frac{\sum_{i=1}^m \left(\hat{y}_i - y_i \right)^2}{\sum_{i=1}^m \left(\bar{y} - y_i \right)^2} \quad (17)$$

$$MAPE = \frac{100\%}{m} \sum_{i=1}^m \frac{\left| \hat{y}_i - y_i \right|}{y_i} \quad (18)$$

$$MAE = \frac{1}{m} \sum_{i=1}^m \left| \hat{y}_i - y_i \right| \quad (19)$$

$$MSE = \frac{1}{m} \sum_{i=1}^m \left(\hat{y}_i - y_i \right)^2 \quad (20)$$

$$RMSE = \sqrt{\frac{1}{m} \sum_{i=1}^m \left(\hat{y}_i - y_i \right)^2} \quad (21)$$

where y_i represents the actual value, $\hat{y}_i$ denotes the predicted value, $\bar{y}$ is the mean value of the actual observations, and m represents the number of data points.

Based on the above five performance evaluation metrics, a CPI was further established to provide an integrated assessment of model performance. The calculation formula is given in Eq. (22). The CPI value ranges from 0.01 to 0.1[29], where a value closer to 0.01 indicates superior predictive performance of the model.

$$CPI = \sqrt[5]{(1 - R^2) \times \frac{MAPE}{100} \times \frac{MAE}{P_{j,\max} - P_{j,\min}} \times \frac{RMSE}{P_{j,\max} - P_{j,\min}} \times \frac{RMSE}{\bar{P}_j}} \quad (22)$$

where P_j represents the value of the j -th performance metric; $P_{j,\min}$ and $P_{j,\max}$ denote the minimum and maximum values of the j -th performance metric among all evaluated models, respectively.

As presented in Table 2, the HPSO-GRU model demonstrates significant superiority over other models across all five classical evaluation metrics and the CPI. Compared with the conventional GRU model and its optimized variants, the proposed model achieves comprehensive improvements in prediction accuracy. Regarding error control, the RMSE of the HPSO-GRU model on the test dataset is reduced by 24% compared with the original GRU model and by 11% compared with the second-

best PSO-GRU model. Similarly, the mean absolute percentage error (MAPE) decreases by 5.5% and 0.6%, respectively. In terms of fitting capability, the HPSO-GRU model achieves the highest coefficient of determination (R^2), indicating its superior ability to capture the nonlinear relationship between input features and temperature variations. Furthermore, the CPI value of the HPSO-GRU model is closest to 0.01, demonstrating the optimal overall prediction performance among all evaluated models. The consistent improvements across multiple performance metrics confirm that the HPSO hybrid optimization algorithm can provide statistically significant and stable performance enhancement for the GRU model when addressing the complex nonlinear regression problem of coal gangue temperature prediction.

Table 2 Performance comparison of different prediction models

Model	Model Evaluation Metrics					
	R^2	MAPE/%	MAE	MSE	RMSE	CPI
GRU	0.9395	11.4206	24.3557	1 066.63	32.66	0.0746
BO-GRU	0.9472	8.1634	22.2816	844.46	29.06	0.0617
IGWO-GRU	0.9518	7.6812	21.2672	795.98	28.21	0.0591
PSO-GRU	0.9613	6.5620	19.8709	777.24	27.88	0.0554
SSA-GRU	0.9411	8.4317	24.0449	996.38	31.57	0.0682
HPSO-GRU	0.9749	5.9164	17.1283	613.38	24.77	0.0464

4.6 Feature Importance

Figure 8 illustrates the feature importance of each input variable, reflecting its contribution to the prediction results[30]. The importance ranking is as follows: $O_2(31.6\%) > CO/\Delta O_2(27.8\%) > CO_2(19.3\%) > CO(17.6\%) > C_2H_4(7.4\%) > C_2H_4/C_2H_6(1.1\%)$. This ranking is highly consistent with the results of the Spearman correlation analysis, indicating that the model prediction is primarily governed by key indicators with clear physical significance. Among these variables, $CO/\Delta O_2$ provides a comprehensive representation of the oxidation reaction intensity, while O_2 concentration directly reflects the degree of oxidation. These findings verify that the proposed model effectively captures the critical mechanistic information associated with the oxidative heating process of coal

gangue rather than overfitting random data fluctuations. Consequently, the interpretability and reliability of the model-based prediction decision-making process are significantly enhanced.

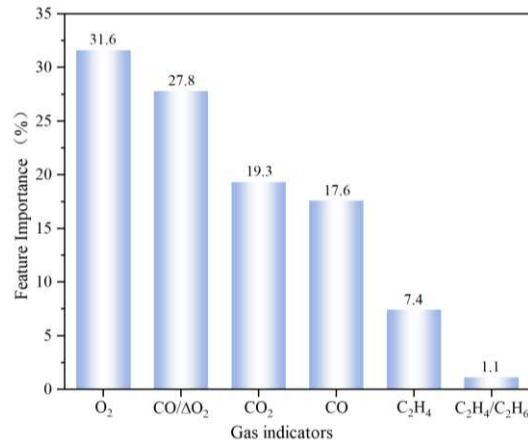


Fig. 8 Feature importance analysis

4.7 Graphical User Interface (GUI) Development

To facilitate the engineering application of the research outcomes, a GUI was developed in MATLAB based on the trained HPSO-GRU model for predicting the internal temperature and oxidative heating stage of coal gangue piles, as shown in Fig. 9. The developed GUI consists of an input module and an output module. Users only need to enter seven characteristic parameters in the input interface and click the “Predict” button to obtain the corresponding temperature prediction results and the associated oxidative heating stage. This provides a convenient and efficient approach for the rapid assessment of spontaneous combustion risks in coal gangue piles.

Input Parameter	Value	Unit
O ₂ volume fraction	20.35	(%)
CO/ΔO ₂	0.12	
CO ₂ concentration	567.6	(ppm)
CO concentration	13.27	(ppm)
C ₂ H ₄ concentration	0	(ppm)
C ₂ H ₄ /C ₂ H ₆	0	
Moisture content	6	(%)

Predict

Output Parameter	Value	Unit
Internal temperature of the coal gangue pile	104	(°C)
Oxidation stage	Slow oxidation	

Fig. 9 GUI for coal gangue internal temperature prediction

5 Conclusions

1) The effect of moisture content on the spontaneous combustion tendency of coal gangue exhibits a nonlinear regulatory mechanism, allowing it to be classified into three zones: an oxidation-promoting zone ($W < 6\%$), a strong oxidation zone ($6\% \leq W \leq 12\%$), and an oxidation-inhibiting zone ($W > 12\%$). Moderate moisture content promotes the dissociation of active functional groups and the release of pore gases, thereby accelerating the oxidation process. However, excessive moisture covers active sites and hinders oxygen diffusion, significantly suppressing spontaneous combustion. Therefore, enhancing the overall water retention capacity of open-air coal gangue piles and maintaining a high-moisture barrier can effectively inhibit oxygen transport and reduce the risk of spontaneous combustion.

2) The prediction performance of the proposed HPSO-GRU model was comprehensively evaluated using the experimental data from the test dataset and compared with those of GRU, BO-GRU, IGWO-GRU, PSO-GRU, and SSA-GRU models. Compared with BO-GRU, SSA-GRU, and GRU, the maximum relative error of the HPSO-GRU model was reduced by 3.6%, 5.1%, and 6.5%, respectively. Moreover, its CPI value was closest to the optimal value of 0.01, indicating superior

overall prediction performance. Furthermore, the feature importance ranking obtained from the proposed model showed high consistency with the Spearman correlation analysis results, thereby enhancing the physical interpretability and reliability of the model outputs.

3) Based on the HPSO-GRU model, a user-friendly graphical interface was developed in MATLAB, enabling rapid prediction of the internal temperature and intelligent identification of oxidation stages in coal gangue piles by inputting six gas indicators and moisture content. The developed system provides an efficient and practical tool for real-time temperature prediction and spontaneous combustion risk assessment of coal gangue piles.

CRedit authorship contribution statement

Peng Wang: Data curation, Methodology, Software, Validation, Visualization, Writing-Original Draft. **Xiuyuan Zhang:** Sampling, Software, writing-review & editing. **Wencai Wang:** Software, Writing-Review & Editing. **Shuang Liang:** Supervision, Writing-Review & Editing. **Shuo Zhang:** Software, Test. **Xiaolei Zhang:** Investigation, Test.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgment

The financial support from the National Natural Science Foundation of China (52464018), The Natural Science Foundation of Inner Mongolia Autonomous Region of china (2025QN05002), Fundamental Research Funds for Inner Mongolia University of Science & Technology (2025QNJS004) is gratefully acknowledged.

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