Effects of Hydrolysis Parameters on AlN Content in Aluminum Dross and Multivariate Nonlinear Regression Analysis

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Abstract: Aluminum dross, as a hazardous waste product, causes harm to the environment and humans, since the AlN it contains chemically reacts with water to produce ammonia. In the present study, a formula for modifying the AlN content in aluminum dross is proposed for the first time, by investigating the components of aluminum dross and changes in their respective contents during the hydrolysis process. Meanwhile, the effects of such hydrolysis parameters as time, temperature, and rotational speed on the hydrolysis rate of aluminum dross are explored. Furthermore, regression analysis is performed on the hydrolysis parameters and objective functions. The results show that as the reaction time increases, the variation in AlN content in aluminum dross decelerates gradually after modification. The hydrolysis rate is the fastest in the initial 4 h, which essentially stagnates after 20 h. The rise in temperature can significantly accelerate the AlN hydrolysis in aluminum dross, while the rotational speed has a non-obvious effect on the hydrolysis rate of AlN in aluminum dross. Regression analysis and secondary simplification are performed on the hydrolysis parameters and the modified AlN content, revealing that the relative error between the theoretical and experimental values is ≤ ±9.34%. The findings of this study have certain guiding significance for predicting and controlling modified AlN content in aluminum dross during hydrolysis.

Keywords: high-nitrogen-content aluminum dross; AlN; hydrolysis parameters; content prediction; modified formula

1. Introduction

Aluminum dross is a waste ash generated during aluminum production, which comes from infusible inclusions [1], oxides, and various additives that float on the aluminum melt surface during smelting [2]. As a typical solid waste product from the aluminum processing industry, it has been included in China’s “National Hazardous Waste List (2021 edition)” due to its reactivity and leaching toxicity [3].

As of today, the only mature technique for utilizing aluminum dross is “ash frying”, where a small amount of elemental aluminum is extracted from aluminum dross to obtain certain economic benefits [4]. However, the extensive remaining ash still cannot be treated effectively, and is often accumulated and buried as solid waste [5], polluting and occupying substantial land resources while seriously affecting the ecological environment and human health [6–8]. Some of the heavy metal elements contained in aluminum dross—such as Hg, Cd, and Pb—can cause severe pollution once they enter the soil or groundwater systems, tremendously impacting the ecosystem.

Additionally, all kinds of aluminum dross contain a certain amount of AlN. To purify the molten aluminum in the smelting process, nitrogen is often filled into the furnace to accelerate the purification and refinement [9]. During the evaporation process, part of the nitrogen is absorbed by the aluminum dross floating on the molten aluminum surface to
form AlN [10]. AlN is a kind of functional ceramic belonging to diamond nitride, which has excellent thermal, mechanical, and electrical properties, including high thermal conductivity (theoretical value 320 W/(m·K)), linear expansivity ($4.8 \times 10^{-6}/K$) comparable to silicon (Si), and outstanding electrical insulation (volume resistivity $> 10^{12} \Omega \cdot m$) [11]. The AlN in aluminum dross tends to be unstable overall due to its formation conditions, structural composition, and refinement difficulty [12]. Under wet or water-exposure conditions, it releases pungent ammonia gas, causing consequences such as imbalance in material performance and changes in solution pH during the utilization process [13]. Additionally, ammonia is a colorless gas with a strong, irritating odor, and can easily cause air pollution due to its strong diffusivity and harmfulness. The ammonia produced by aluminum dross hydrolysis is highly toxic to the human body [14]. Chronic intoxication can cause such respiratory diseases as bronchitis and emphysema, while acute intoxication can cause severe consequences such as persistent coughing, suffocation, and even death [15].

Clearly, the landfill of aluminum dross and related pollution have become hot issues in today’s social development, and go against the current sustainability concept of zero emissions and pollution. The requirements of reducing, preventing, treating, and upcycling waste are imminent. There are various experimental methods for AlN, such as cathodoluminescence measurements (CL), X-ray absorption near-edge spectroscopy (XANES), and Fourier-transform infrared spectroscopy (FTIR) [16–18]. This study systematically analyzes the AlN (in aluminum dross) hydrolysis parameters such as temperature, time, and rotational speed, and performs their selection and optimization. Meanwhile, on the basis of extensive experimental data, each hydrolysis parameter and objective function (AlN content) is regressively analyzed, in order to achieve prediction and control of AlN in aluminum dross.

2. Materials and Methods
2.1. Raw Material and Pretreatment

The ultimate aluminum dross used in this experimentation came from a secondary aluminum company in Jiangsu, China, and was analyzed and tested after pretreatment. The pretreatment process was as follows: The aluminum dross was preliminarily screened via a standard large-mesh sieve to remove the aluminum metal blocks. Then, the remainder was ground, and screened using a standard small-mesh sieve to yield the experimental raw material [19]. Table 1 details the XRD results of the aluminum dross.

| Component | Al  | Al$_2$O$_3$ | AlN  | AlO(OH) | SiO$_2$ |
|-----------|-----|-------------|------|---------|---------|
| Content (%) | 5 ± 2 | 20 ± 2 | 13 ± 2 | 3 ± 1 | 1       |

2.2. Reagents and Instruments

The reagents used in the experimentation were NaOH (analytically pure, Xilong Scientific Co., Ltd., Shenzhen, China), methyl red (indicator, Yuanye Biotechnology), methylene blue (indicator, COOLABER SCIENCE & TECHNOLOGY), HCl (analytically pure, Lingfeng Chemical Reagent, Shanghai, China), and boric acid (chemically pure, Meryer Chemical Technology, Shanghai, China). Experimental instruments were as follows: ultra water purifier (EPED-10TH, EPED Technology, Nanjing, China), universal electric furnace (DK-98 type, Chushui Electrothermal Appliance Co., Ltd., Chushui, China), digital stirrer (JB60-5H, Lichen Instrument, Shaoxing, China), water bath (WB-4, Zhengrong Experimental Instrument), and electro-thermostatic blast oven (101A-2, Hengchang Instrument, Haimen, Nantong, China).
2.3. Analytical Test Methods

The aluminum dross was hydrolyzed, filtered, and dried under different hydrolysis parameters, and a distillation separation unit was built by utilizing the aforementioned instruments. Figure 1 illustrates the schematic of the unit.

![Figure 1. Schematic diagram of distillation separation plant (1—electric furnace; 2—asbestos net; 3,5—cone bottle; 4—condenser tube).](image)

After weighing 2 g of aluminum dross, it was added to an Erlenmeyer flask containing 150 mL of 20%wt NaOH solution. The flask was quickly stoppered, and then heated and kept boiling for approximately 2 h to allow for distillation. Meanwhile, the distilled ammonia gas was absorbed with 200 mL of 40 g/L boric acid solution. Following distillation, titration was performed with 0.05 mol/L dilute HCl solution, where the standard methyl red–methylene blue served as an indicator. The endpoint was marked by a blue-to-purple change in solution color [20,21].

A blank experiment was conducted according to the aforementioned procedure, and the relative volume of HCl consumed in the experiment was determined, based on which the AlN content was calculated as follows:

\[
Z = \frac{0.041C(V_2 - V_1)}{M} \times 100\%
\]

where \(C\) denotes the concentration of dilute HCl (mol/L), \(V_2\) and \(V_1\) represent the volumes of diluted HCl consumed in the titration process and the blank experiment, respectively (mL), \(M\) denotes the sample mass (g), and \(Z\) denotes the AlN content in aluminum dross directly measured after hydrolysis (%).

3. Effects of Hydrolysis Parameters on the Nitrogen Removal Efficiency from Aluminum Dross

3.1. Prediction of AlN Content and Its Modification Formula

The AlN contents in different aluminum dross varied distinctly. To validate the accuracy of subsequent tests, predicting the AlN content in hydrolyzed products was necessary. Figure 2 depicts the XRD spectra of aluminum dross. Clearly, the hydrolyzed products contained Al, \(\text{Al}_2\text{O}_3\), AlN, and other crystalline substances (amorphous components were excluded from the test results). The semi-quantitative XRD results of aluminum dross are detailed in Table 2. Since the aluminum dross used herein was already processed by ash frying, the AlN content was low, while the contents of \(\text{Al}_2\text{O}_3\) and AlN were about 20% and 10%, respectively. Figure 3 displays the local EDS spectra at two material sites. Since almost all nitrogen contained in the aluminum dross was AlN, after conversion, the AlN contents in Figure 3a,b were estimated to be 12.97% and 16.37%, respectively.
The above analysis revealed that the main components of hydrolyzed products and their respective contents conformed to the definition of "secondary aluminum dross" in the previous paper, and that the hydrolyzed products were the dross residues from ash frying, showing good agreement with the actual situation. The AlN contents in the hydrolyzed products were predicted to range between 10 and 17%.

The hydrolytic reaction of AlN in aluminum dross is described in Equation (2). During this process, the N in AlN was separated from the aluminum dross in the form of NH$_3$, and its elemental position was occupied by OH$^{-}$ ions to form Al(OH)$_3$ with a molecular mass of 78, which was considerably greater than the original AlN’s 41. Additionally, apart from insoluble substances such as Al and Al$_2$O$_3$, the aluminum dross also contained small amounts of soluble salts, such as NaCl and KCl [22,23].

$$\text{AlN} + 3\text{H}_2\text{O} = \text{Al(OH)}_3 + \text{NH}_3$$

$$41 : 54 = 78 : 17 \quad (2)$$

If the above determinations were used directly as the AlN contents in aluminum dross, not only would the changes in other components and contents be ignored, but the AlN content standard also could not be unified. In particular, when measuring AlN-rich aluminum dross, the error would be larger. To address this problem, the compositional and content variations during the hydrolysis process in Equation (2) were investigated, thereby deriving the following formula:

$$Z = \frac{KM - m}{M - Y + \frac{28}{41}m} \quad (3)$$
3.2. Effects of Time and Rotational Speed on the AlN Content

Under different rotational speeds, the aluminum dross was hydrolyzed at 60 °C. Figure 4 depicts the AlN variation trends in the aluminum dross after modification. Clearly, under mechanical stirring at both low speeds (0, 100, and 300 r/min) and high speeds (600 and 1000 r/min), the variation trends of modified AlN contents remained generally consistent. Moreover, at various time points, the modified AlN contents in aluminum dross differed little, suggesting an insignificant effect of rotational speed on the aluminum dross hydrolysis rate.

Meanwhile, the modified AlN content in aluminum dross decreased gradually with the increasing hydrolysis time, decreasing by about half (from 12.68% to 6.50%) under this hydrolysis condition 24 h after reaction. More obviously, at the 0–4 h stage, the modified
AIN content decreased drastically from the initial 12.68% to 9.00–10.40%, showing considerably sharper reduction than that at other reaction time periods. With the prolongation of hydrolysis time, the aluminum dross hydrolysis rate was gradually lowered, and the variation trends of modified AlN contents in aluminum dross were generally consistent after 20 h.

![Figure 4](image_url)

**Figure 4.** The modified contents of AlN in aluminum dross under different rotational speeds (60 °C).

The causes of the above phenomenon were as follows: On the one hand, the particle size and surface area of aluminum dross were large at the initial reaction stage, and the water contact area was also correspondingly large. Since more AlN participated in the reaction under identical hydrolysis conditions, the hydrolytic process was faster. On the other hand, given the basic hydrolysis properties of AlN in aluminum dross, the reaction product Al(OH)$_3$ was a white insoluble gelatinous precipitate. With the prolongation of hydrolysis time, this product and its attachments gradually blocked the pores of aluminum dross particles, which hindered the contact channel between AlN and water, ultimately affecting the hydrolysis rate of aluminum dross. Meanwhile, stirring facilitated the suspension of aluminum dross particles in the hydrolysis system, prevented the accumulation of dross at the system’s bottom, and promoted the diffusion of generated NH$_3$ outside the hydrolysis system. However, at identical times and temperatures, the rotational speed generally did not affect the AlN–water contact area in the aluminum dross, which also had only a small effect on the hydrolysis process.

### 3.3. Effects of Temperature on the AlN Content

Under different temperature conditions, the aluminum dross hydrolysis experiments were carried out at 600 r min$^{-1}$. Figure 5 illustrates the relevant variation trends. As is clear, at identical hydrolysis times, the modified AlN content in aluminum dross was markedly lower in the hydrolysis system at 100 °C than that at 60 °C and 80 °C. After 24 h of hydrolysis, the modified AlN content at a 100 °C hydrolysis temperature decreased from the original 12.88% to 2.25%, and there was no obvious ammonia smell upon further continuation of the hydrolysis. Meanwhile, after 24 h of hydrolysis at 80 °C, the modified AlN content dropped to 4.23%, and ammonia was slightly smelt upon further hydrolysis. In contrast, at 60 °C, the AlN content only dropped to 6.28%, and apparent bubbles and ammonia smell were produced upon further hydrolysis, indicating that the hydrolysis process had not ended at 60 °C. Clearly, the increase in temperature could remarkably promote the hydrolysis of aluminum dross. It could be speculated that with the prolongation...
of hydrolysis time, the variation of modified AlN content in aluminum dross gradually slowed down, so it inevitably took a long time to achieve complete hydrolysis.

Figure 5. The modified contents of AlN in aluminum dross under different temperatures and speeds (600 r·min\(^{-1}\)).

The rate of a chemical reaction varies by temperature. The effect of temperature on the reaction rate is manifested primarily in the rate coefficient of reaction. Normally, the higher the temperature, the greater the reaction rate coefficient, which is directly manifested by the acceleration of the reaction rate [24]. At the end of the 19th century, the Swedish chemist Arrhenius proposed the famous Arrhenius equation after investigating the relationship between the hydrolysis rate of sucrose and the temperature:

\[
\ln(k) = \ln(k_0) - \frac{E_a}{RT} \tag{5}
\]

where \(k\) represents the reaction rate coefficient (dimensionless), \(k_0\) represents the pre-exponential parameter (dimensionless), \(E_a\) denotes the reaction activation energy (kJ·mol\(^{-1}\)), \(T\) denotes the temperature (K), and \(R\) is the molar gas constant (J (mol·K)\(^{-1}\)).

Both \(k_0\) and \(E_a\) are empirical parameters, which can be regarded as temperature-independent constants if the temperature fluctuates within a small range. At this time, the Arrhenius equation has excellent applicability. In different-temperature reaction systems, the correlation between the rate coefficients at two varying temperatures could be derived by Equation (5):

\[
\ln(k_1) = \ln(k_0) - \frac{E_a}{RT_1} \tag{6}
\]

\[
\ln(k_2) = \ln(k_0) - \frac{E_a}{RT_2} \tag{7}
\]

Supposing that \(T_1 < T_2\) within an interval \(T_1 – T_2\), and that \(k_0\) and \(E_a\) could be deemed as constants, Equation (8) could be derived based on Equations (6) and (7). Since \(E_a > 0\) and \(T_1 < T_2\), the right side of Equation (8) was greater than 0, so \(k_2/k_1 > 1\). In other words, the reaction rate coefficient at high temperatures was generally greater than that at low temperatures. Hence, the rise in temperature could increase the hydrolysis rate of AlN in the aluminum dross.

\[
\ln \left( \frac{k_2}{k_1} \right) = \frac{E_a}{R} \left( \frac{T_2 - T_1}{T_1T_2} \right) \tag{8}
\]
Additionally, the microscopic essence of reaction lay in the breakage of old chemical bonds and the formation of new chemical bonds caused by effective particle collision. The rise in temperature increased the percentage and motion speed of activated molecules, and the increase in the number of activated molecules further promoted the effective intermolecular collision. Accordingly, the numbers of broken old bonds and formed new bonds per unit of time also increased.

To sum up, the hydrolysis rate of AlN in aluminum dross could be effectively accelerated by increasing the hydrolysis temperature and prolonging the hydrolysis time. As the hydrolysis time increased, the AlN hydrolysis rate gradually decreased, and after 20 h, the hydrolytic process essentially stopped. Meanwhile, the rotational speed produced a non-obvious effect on the hydrolysis rate of AlN in aluminum dross.

### 3.4. Effects of Hydrolysis Conditions on the pH

#### 3.4.1. N Migration Model of Aluminum Dross during Hydrolysis

The N-containing compound in the aluminum dross is mostly AlN, and all of the N during the hydrolysis process is generally believed to come from this phase. After in-depth research and analysis, we proposed the N migration model of aluminum dross during its hydrolysis, as shown in Figure 6.

![Figure 6. N migration model of aluminum dross during hydrolysis.](image)

On the one hand, the NH$_3$ produced during the hydrolysis of AlN in aluminum dross was dissolved in suspension as a soluble gas, and then hydrated to form NH$_3$·H$_2$O. On the other hand, part of the undissolved NH$_3$ diffused above the suspension. Moreover, like other gas–liquid two-phase processes, the gas and liquid phases were dynamically interacting rather than mutually isolated. In macroscopic terms, the NH$_3$·H$_2$O in suspension was decomposed by heat, so that the ammonia gas was diffused above the suspension. Meanwhile, the ammonia gas in the air above the suspension was also continuously dissolved into the suspension. In the meantime, the formed NH$_3$·H$_2$O ionized into NH$_4^+$ and OH$^-$, which also continued to reversely combine to form NH$_3$·H$_2$O [25,26]. The above reactions interacted to establish a stable dynamic equilibrium system. By measuring the pH of the suspension in the equilibrium state, and referring to the “direct characterization” procedure for measuring the AlN content in aluminum dross, we assessed the feasibility of the “indirect characterization” procedure for determining the hydrolysis rate of aluminum dross.

#### 3.4.2. Effects of Time and Rotational Speed on the Suspension pH

Figure 7 depicts the variation trends of the suspension pH under different rotational speeds at 60 °C. At the 0–2 h reaction stage, the suspension pH values under various rotational speeds increased sharply. With the prolongation of reaction time, the suspension pH values at low rotational speeds (0 and 100 r min$^{-1}$) were significantly greater than those at high rotational speeds (300 and 600 r min$^{-1}$). The primary cause of this phenomenon was that under identical conditions, a low rotational speed was conducive to the NH$_3$ retention in the suspension, which led to a higher concentration of OH$^-$ ions ionized from the formed NH$_3$·H$_2$O, so that the pH value of the suspension was higher. Additionally, neither low
nor high rotational speeds produced particularly obvious effects on the suspension pH. Overall, the rotational speed and the suspension pH appeared to be uncorrelated.

\[ K_1 = \frac{C(NH_4^+)}{C(NH_3 \cdot H_2O)} \cdot \frac{C(OH^-)}{C(NH_3 \cdot H_2O)} = \frac{C^2(OH^-)}{C(NH_3 \cdot H_2O)} \]  \hspace{1cm} (9)

\[ K_2 = \frac{C(H^+)}{C(H_2O)} \cdot \frac{C(OH^-)}{C(H_2O)} \]  \hspace{1cm} (10)

where \( K_1 \) represents the ionization constant of ammonia water (dimensionless), \( K_2 \) represents the ionization constant of water (dimensionless), \( C(NH_4^+) \) denotes the concentration of \( NH_4^+ \) ionized from ammonia water (mol L\(^{-1}\)), \( C(OH^-) \) denotes the concentration of \( OH^- \) ionized from ammonia water (mol L\(^{-1}\)), \( C(H^+) \) denotes the concentration of \( H^+ \) ionized from water (mol L\(^{-1}\)), \( C(NH_3 \cdot H_2O) \) is the concentration of unionized \( NH_3 \cdot H_2O \) in the solution (mol L\(^{-1}\)), and \( C(H_2O) \) is the concentration of unionized \( H_2O \) in the solution (mol L\(^{-1}\)).
Combining Equations (9) and (10) gives:

\[
\text{pH} = -\log(CH^+) = \log\frac{K_1 \cdot C(NH_3 \cdot H_2O)}{K_2 \cdot C(H_2O)}
\]  

(11)

As is clear from Equation (11), assuming that the reaction system was at the saturation stage of ammonia, the \(C(NH_3 \cdot H_2O)\) and \(C(H_2O)\) values in the system were constant, and the pH value of the suspension was related mainly to the ammonia ionization constant \(K_1\) and the water ionization constant \(K_2\). Since these two constants were directly correlated with the temperature \(T\), the resultant values were essentially nonlinear, so the suspension pH could not accurately characterize the hydrolysis rate of aluminum dross under different temperature conditions. Meanwhile, according to Figure 8, when “indirect measures” such as suspension pH were used to assess the hydrolysis rate of aluminum dross, it was impossible to intuitively comprehend the effects of various parameters on the dross hydrolysis rate.

4. Regression Analysis of Hydrolysis Parameters

Based on the above experiments, during the aluminum dross hydrolysis, temperature, rotational speed, and time were independent variables that affected the AlN content (dependent variable). Since linear regression is a type of nonlinear regression, and there were multiple variables in the present experimentation, the “Analysis” module in Origin was utilized to perform relevant nonlinear regression analysis based on ternary quadratic polynomials. The preliminary regression model was formulated as follows:

\[
y = a x_1^2 + bx_2^2 + cx_3^2 + dx_1 x_2 x_3 + ex_1 x_2 + f x_1 x_3 + g x_2 x_3 + hx_1 + ix_2 + j x_3 + k
\]  

(12)

where \(x_1\) denotes the hydrolysis temperature of aluminum dross (°C), \(x_2\) denotes the hydrolysis speed of aluminum dross (r min\(^{-1}\)), \(x_3\) denotes the hydrolysis time of aluminum dross (h), \(y\) represents the modified content of AlN (%), and others were undetermined constants (dimensionless).

As suggested by the above analysis, the independent variable “rotational speed” had an insignificant effect on the variation of AlN content in aluminum dross. To simplify the model, the rotational speed \(x_2\) was “zeroed”. The simplified regression model is as follows:

\[
y = a x_1^2 + c x_3^2 + e x_1 x_2 + f x_1 x_3 + h x_1 + j x_3 + k
\]  

(13)

Figure 8. Suspension pH values at different temperatures (300 r min\(^{-1}\)).
Table 3 lists the values of various regression coefficients in Equations (12) and (13). The coefficient of multiple determination $R^2$, F-statistic, and $p$-value were $0.93035$, $508.32976$, and $0$ in the preliminary model, respectively, while they were $0.93616$, $1016.49681$, and $0$ in the simplified model, respectively. This indicates that the correlations between dependent and independent variables were higher with the simplified model, which yielded more significant results and achieved a better fitting effect [27,28].

Table 3. Numerical statistics of regression model.

| Regression Coefficients | The Values of the Regression Coefficients | Standard Deviation of Regression Coefficient |
|-------------------------|------------------------------------------|--------------------------------------------|
|                         | Preliminary Model | Simplified Model | Preliminary Model | Simplified Model |
| a                       | 0.00122 | 9.57857 × 10^{-4} | 8.38698 × 10^{-4} | 7.3811 × 10^{-4} |
| b                       | −1.36763 × 10^{-6} | — | 1.24974 × 10^{-6} | — |
| c                       | 0.01812 | 0.01812 | 0.00193 | 0.00185 |
| d                       | 0.27207 | — | — | — |
| e                       | 0.2718 | — | — | — |
| f                       | −163.24244 | −0.00267 | — | 8.8037 × 10^{-4} |
| g                       | −16.32398 | — | — | — |
| h                       | −163.32872 | −0.20235 | — | 0.12209 |
| i                       | −16.30633 | — | — | — |
| j                       | 9793.85899 | −0.52673 | — | 0.07604 |
| k                       | 9807.86654 | 21.45126 | — | 4.61293 |

Preliminary model: $R^2 = 0.93035$, F = 508.32976, $p = 0$; preliminary model: $R^2 = 0.93616$, F = 1016.49681, $p = 0$

The preliminary model is specifically expressed as follows:

$$
yy = 0.00122x_1^2 - 1.36763 \times 10^{-6}x_2 + 0.01812x_3^2 + 0.27207x_1x_2x_3
\quad + 0.2718x_1x_2 - 163.24244x_1x_3
\quad - 16.32398x_2x_3 - 163.32872x_1 - 16.30633x_2
\quad + 9793.85899x_3 + 9807.86654
$$

(14)

The simplified model is specifically expressed as follows:

$$
y = 9.57857 \times 10^{-4}x_1^2 + 0.01812x_3^2 - 0.00267x_1x_3 - 0.20235x_1 - 0.52673x_3 + 21.45126
$$

(15)

Table 4 compares the experimental and theoretical values between the preliminary and simplified models. Clearly, the simplified treatment could maintain good accuracy, with relative errors controlled generally within ±9.34%, which could somewhat guide the prediction and control of the AlN content in aluminum dross during hydrolysis.

Table 4. Comparison between experimental modified values and theoretical modified values of the AlN hydrolysis process in aluminum dross (part).

| Number | Experimental Value | Theoretical Value | Relative Error (%) |
|--------|--------------------|-------------------|--------------------|
|        |                    | Preliminary Model | Simplified Model    |
|        |                    | Preliminary Model | Simplified Model    |
| 1      | 7.07               | 6.53              | −7.64              | −9.34              |
| 2      | 6.86               | 6.83              | −0.44              | −2.19              |
| 3      | 12.88              | 12.97             | 0.70               | −0.93              |
| 4      | 4.59               | 4.16              | −9.37              | −8.71              |
| 5      | 4.15               | 3.83              | −7.71              | −7.71              |
| 6      | 4.23               | 4.06              | −4.02              | −4.02              |
| 7      | 7.53               | 7.07              | −6.11              | −5.44              |
| 8      | 3.82               | 3.88              | 1.57               | 1.57               |
| 9      | 2.25               | 2.18              | −3.11              | −3.11              |
5. Conclusions

(1) By investigating the components of aluminum dross and changes in their respective contents during the hydrolysis process, a formula for modifying the AlN content in aluminum dross is proposed for the first time. Capable of reflecting the absolute content of AlN in aluminum dross, the formula provides a new way to scientifically characterize the hydrolysis rate of aluminum dross.

(2) The experimental study of aluminum dross hydrolysis under different times, temperatures, and rotational speeds found that after 24 h of hydrolysis, the modified AlN content decreased from the original 12.88% to 2.25% at a 100 °C hydrolysis temperature, dropped to 4.23% at 80 °C, and only dropped to 6.28% at 60 °C. The hydrolytic process essentially stopped 20 h later. The aluminum dross hydrolysis can be effectively promoted by the prolongation of time and the increase in temperature. Meanwhile, rotational speed produces an insignificant effect on the dross hydrolysis process.

(3) Through the hydrolysis experiments under different conditions, we found that the suspension pH increased sharply during the 0–2 h period. With the prolongation of reaction time, the suspension pH decreased slowly, exhibiting greater values at low rotational speeds (0 and 100 r min⁻¹) than at high rotational speeds (300 and 600 r min⁻¹). Meanwhile, the rise in temperature can significantly lower the maximum pH of the suspension in the system.

(4) Comparison of two methods for characterizing aluminum dross hydrolysis rate—namely, the AlN content detection, and the suspension pH measurement—reveals that the suspension pH is greatly affected by temperature and NH₃ release. Moreover, the results obtained at identical temperatures are obviously inconsistent with the direct characterization results. Overall, the direct detection of AlN content in aluminum dross is versatile and highly accurate.

(5) Multivariate nonlinear regression was performed between the modified AlN content and hydrolysis parameters such as time, temperature, and rotational speed. After removing the outliers in the model, the hydrolysis parameters and AlN content were subjected to multivariate nonlinear regression and quadratic simplification to derive a simplified model. With the simplified model, the relative errors between the theoretical and experimental values of modified AlN content were within ±9.34%, which can somewhat guide the prediction and control of AlN content in aluminum dross during hydrolysis.

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References

1. Foo, C.T.; Salleh, M.A.M.; Ying, K.K.; Matori, K.A. Mineralogy and thermal expansion study of mullite-based ceramics synthesized from coal fly ash and aluminum dross industrial wastes. *Ceram. Int.* 2019, 45, 7488–7494. [CrossRef]

2. Choo, T.F.; Mohd Salleh, M.A.; Kok, K.Y.; Matori, K.A.; Abdul Rashid, S. Characterization of high-temperature hierarchical Porous Mullite washcoat synthesized using Aluminum Dross and Coal fly ash. *Crystals* 2020, 10, 178. [CrossRef]
3. Elseknidy, M.H.; Salmiaton, A.; Nor Shafizah, I.; Saad, A.H. A Study on Mechanical Properties of Concrete Incorporating Aluminum Dust. Fly Ash, and Quarry Dust. *Sustainability* **2020**, *12*, 9230. [CrossRef]

4. Kikuchi, R. Recycling of municipal solid waste for cement production: Pilot-scale test for transforming incineration ash of solid waste into cement cinder. *Resour. Conserv. Recycl.* **2001**, *31*, 137–147. [CrossRef]

5. Mohammadzadeh, K.; Mahinroosta, M.; Allahverdi, A.; Dong, P.; Bassim, N. Non-supercritical drying synthesis and characterization of monolithic alumina aerogel from secondary aluminum dross. *Ceram. Int.* **2022**, *48*, 13154–13162. [CrossRef]

6. Ni, H.; Zhang, J.; Lv, S.; Gu, T.; Wang, X. Performance Optimization of Original Aluminum Ash Coating. *Coatings* **2020**, *10*, 831. [CrossRef]

7. Mailar, G.; Sreedhara, B.M.; Manu, D.S.; Hiremath, P.; Jayakesh, K. Investigation of concrete produced using recycled aluminum dross for hot weather concreting conditions. *Resour.-Effic. Technol.* **2016**, *7*, 68–80. [CrossRef]

8. Lee, J.R.; Lee, I.; Shin, H.Y.; Ahn, J.G.; Kim, D.J.; Chung, H.S. Nitride-related compounds preparation from waste aluminum dross by self-propagating high-temperature process. In *Materials Science Forum*; Trans Tech Publications Ltd.: Schwyz, Switzerland, 2005; Volume 486, pp. 297–300. [CrossRef]

9. Lu, M.; Abidin, Z.; Riani, E.; Ismail, A. Utilization of hazardous waste of black dross aluminum: Processing and application-a review. *J. Degrada. Min. Lands Manag.* **2022**, *9*, 3265–3271. [CrossRef]

10. Zhang, F.; Zhang, J.; Zhu, Y.; Wang, X.; Jin, Y. Microstructure and Properties of Polytetrafluoroethylene Composites Modified by Carbon Materials and Aramid Fibers. *Coatings* **2020**, *10*, 1103. [CrossRef]

11. Sooksaen, P.; Puathawee, P. Properties of Unglazed Ceramics Containing Aluminum Dross as a Major Component. In *Solid State Phenomena*; Trans Tech Publications Ltd.: Schwyz, Switzerland, 2017; Volume 266, pp. 182–186. [CrossRef]

12. Benkhelif, A.; Kolli, M. Synthesis of Pure Magnesium Aluminate Spinel (MgAl<sub>2</sub>O<sub>4</sub>) from Waste Aluminum Dross. *Waste Biomass Valorization* **2012**, *3*, 2637–2649. [CrossRef]

13. Kale, M.; Yilmaz, I.H.; Kaya, A.; Çetin, A.E.; Söylemez, M.S. Pilot-scale hydrogen generation from the hydrolysis of black aluminum dross without any catalyst. *J. Energy Inst.* **2022**, *100*, 99–108. [CrossRef]

14. Tang, J.; Liu, G.; Qi, T.; Zhou, Q.; Peng, Z.; Li, X.; Yan, H.; Hao, H. Two-stage process for the safe utilization of secondary aluminum dross in combination with the Bayer process. *Hydrometallurgy* **2022**, *209*, 105836. [CrossRef]

15. Guo, J.; Zhou, Z.; Ming, Q.; Huang, Z.; Zhu, J.; Zhang, S.; Xu, J.; Xi, J.; Zhao, Q.; Zhao, X. Recovering precipitates from dechlorination process of saline wastewater as Poly aluminum chloride. *Chem. Eng. J.* **2022**, *427*, 131612. [CrossRef]

16. Bellucci, S.; Popov, A.I.; Balasubramanian, C.; Cinque, G.; Marcelli, A.; Karbovnyk, I.; Savchyn, V.; Krutyak, N. Luminescence, vibrational and XANES studies of AlN nanomaterials. *Radiat. Meas.* **2007**, *42*, 708–711. [CrossRef]

17. Balasubramanian, C.; Bellucci, S.; Castrucci, P.; De Crescenzi, M.; Bhoraskar, S. Scanning tunneling microscopy observation of coiled aluminum nitride nanotubes. *Chem. Phys. Lett.* **2004**, *383*, 188–191. [CrossRef]

18. Zhukovskii, Y.F.; Pugno, N.; Popov, A.I.; Balasubramanian, C.; Bellucci, S. Influence of F centres on structural and electronic properties of AlN single-walled nanotubes. *J. Phys. Condens. Matter* **2007**, *19*, 395021. [CrossRef]

19. Shuaishuai, L.; Jiaqiao, Z.; Hongjun, N.; Xingxing, W.; Yu, Z.; Tao, G. Study on Preparation of Aluminum Ash Coating Based on Plasma Spray. *Appl. Sci.* **2019**, *9*, 4980.

20. Ni, H.; Zhang, J.; Lv, S.; Wang, X.; Zhu, Y.; Gu, T. Preparation and Performance Optimization of Original Aluminum Ash Coating Based on Plasma Spraying. *Coatings* **2019**, *9*, 770. [CrossRef]

21. Zhang, F.; Zhang, J.; Ni, H.; Zhu, Y.; Wang, X.; Wan, X.; Chen, K. Optimization of AlSi<sub>10</sub>MgMn Alloy Heat Treatment Process Based on Orthogonal Test and Grey Relational Analysis. *Crystals* **2021**, *11*, 385. [CrossRef]

22. Zhang, Y.; Ni, H.; Lv, S.; Wang, X.; Li, S.; Zhang, J. Preparation of Sintered Brick with Aluminum Dross and Optimization of Process Parameters. *Coatings* **2021**, *11*, 1039. [CrossRef]

23. Calignano, F.; Manfredi, D.; Ambrosio, E.P.; Iuliano, L.; Fino, P. Influence of process parameters on surface roughness of aluminum parts produced by DMLS. *Int. J. Adv. Manuf. Technol.* **2013**, *67*, 2743–2751. [CrossRef]

24. Felix, G.S.; Sellin, N.; Marangoni, C. Reduction of dross in galvanized sheets through automatic control of snout positioning in continuous operation. *Int. J. Adv. Manuf. Technol.* **2017**, *89*, 2345–2353. [CrossRef]

25. Zhou, C.; Wang, Q.; Zhang, W.; Zhao, W. Recovery of ammonia nitrogen from aluminum slag ash. *Conserv. Util. Miner. Resour.* **2012**, *3*, 38–41. [CrossRef]

26. Yoldı, M.; Fuentes-Ordoñez, E.G.; Korili, S.A.; Gil, A. Efficient recovery of aluminum from saline slag wastes. *Miner. Eng.* **2019**, *140*, 105884. [CrossRef]

27. Yang, Q.; Li, Q.; Zhang, G.; Shi, Q.; Feng, H. Investigation of leaching kinetics of aluminum extraction from secondary aluminum dross with use of hydrochloric acid. *Hydrometallurgy* **2019**, *187*, 158–167. [CrossRef]

28. Shabashov, V.A.; Kozlov, K.A.; Lyashkov, K.A.; Litvinov, A.V.; Dorofeev, G.A.; Titova, S.G.; Fedorenko, V.V. Effect of aluminum on mechanical solid-state alloying of iron with nitrogen in ball mill. *Phys. Met. Metallogr.* **2012**, *113*, 992–1000. [CrossRef]