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Effects of Sodium Carbonate and Calcium Oxide on Roasting Denitrification of Recycled Aluminum Dross with High Nitrogen Content

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Abstract: Aluminum dross is solid waste produced by the aluminum industry. It has certain toxicity and needs to be treated innocuously. The effect of sodium carbonate and calcium oxide on the denitrification efficiency of high nitrogen aluminum dross roasting was studied in this paper. By means of XRD, SEM and other characterization methods, the optimum technological parameters for calcination denitrification of the two additives were explored. The test results show that both additives can effectively improve the efficiency of aluminum dross roasting denitrification, and the effect of sodium carbonate is better. When the mass ratio of sodium carbonate to aluminum dross is 0.6, the roasting temperature is 1000 °C and the roasting time is 4 h, the denitrification rate can reach 91.32%.

Keywords: roasting denitrification; additive; process parameter

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1. Introduction

Aluminum dross is solid waste produced in the process of aluminum production. It is mainly composed of non-molten impurities, oxides and various additives floating on the surface of aluminum melt during smelting [1]. According to the source of aluminum dross, aluminum dross can be divided into primary aluminum dross and secondary aluminum dross. Primary aluminum dross usually refers to the scum insoluble in aluminum liquid generated in the process of electrolytic aluminum and cast aluminum. The content of metallic aluminum in primary aluminum dross is high up to 30–70%. In addition, it also contains fluorinated salt, aluminum oxide, aluminum nitride and other substances. The color is generally gray-white, also known as white aluminum dross. Secondary aluminum dross usually refers to the waste residue generated in secondary aluminum industry processes such as remelting primary aluminum dross or recovering aluminum alloy from waste aluminum. A variety of salt solvents need to be added in the process of recovering metal aluminum in the secondary aluminum industry. Therefore, the composition of secondary aluminum dross is more complex than that of primary aluminum dross, mainly including metal aluminum, aluminum oxide, aluminum nitride, aluminum carbide, chloride, etc. The content of metal aluminum is less, which can be reduced to less than 10%. The secondary aluminum dross is generally black, also known as black dross.

In 2020, the global output of recycled aluminum will reach 34.71 million tons, accounting for 33.10% of the total amount of primary aluminum and recycled aluminum [2]. The output of recycled aluminum in developed countries has generally exceeded the output of primary aluminum, such as the United States, which has accounted for more than 80% of the total. While the recycled aluminum industry provides aluminum materials for economic construction, a large amount of aluminum dross, namely recycled aluminum dross, is also produced in the regeneration process [3]. Recycled aluminum dross generally belongs to secondary aluminum dross. There are few heavy metals in aluminum dross slag, but there are still heavy metals such as Se, Cr and Pb [4]. A large amount of accumulation will cause heavy metal pollution of surrounding soil and groundwater [5]. Soluble salts such as NaCl and KCl contained in aluminum dross will enter the soil with rainwater and cause soil salinization [6]. Fluoride in aluminum dross easily causes fluoride pollution, resulting in muddy soil when it is wet. At the same time, aluminum dross contains many chemical reactive substances, which makes it dangerous and toxic. For example, AlAs in aluminum dross will react with water to produce AsH_3 gas, which will lead to hydrogen arsenide poisoning of close contacts and endanger the health of nearby personnel [7]. Metal aluminum and aluminum carbide will react with water to produce H_2 and CH_4 combustible gases and release a large amount of heat, which is easy to cause a fire [8]. In particular, aluminum dross is rich in AlN, which can react with water in the air and slowly release the unpleasant irritant gas NH_3, seriously polluting the atmosphere [9]. Human beings have a very sensitive sense of smell for ammonia. The concentration of ammonia in the air reaches 15.2 mg/m^3, and human beings have an obvious sense of stimulation [10]. When the ammonia concentration is 1290.2 mg/m^3, humans can cause severe cough, seriously damage the respiratory system and may die within 30 min [11]. Therefore, aluminum dross is recognized as hazardous waste in the National Hazardous Wastes Catalogue (2021 Edition) [12] and the European Hazardous Waste List [13].

At present, the treatment methods for recycled aluminum dross are mainly stockpiling and landfill [14], which will cause irreparable damage to the environment. Therefore, it is necessary to find a harmless treatment method with low cost and high efficiency to realize the harmless treatment of recycled aluminum dross. The content of fluoride in recycled aluminum dross is low, and its properties are relatively stable, which will not affect the performance of resource utilization products, and even fluoride can be removed in the process of resource utilization [15]. Recycled aluminum dross contains a large amount of aluminum nitride, and its properties are very unstable [16]. However, the existing aluminum dross recycling technology cannot directly remove AlN, so the recycled aluminum dross needs to be pretreated for nitrogen removal during the recycling process [17].

This paper aims to explore the effect of sodium carbonate and calcium oxide on the denitrification efficiency of aluminum dross roasting. The effects of additive ratio, calcination temperature and calcination time on the denitrification effect of high nitrogen content aluminum dross were studied, and the best process parameters were found to provide a reference for the study of the harmless treatment process of aluminum dross.

2. Experiment

2.1. Raw Material, Reagent and Instrument

Raw material: Recycled aluminum dross with high nitrogen concentration comes from Jiangsu Haiguang Metal Co., Ltd. (Suqian, China), which is the aluminum dross with high nitrogen concentration obtained after recycling part of metal aluminum with aluminum dross. The production flow chart is shown in Figure 1.

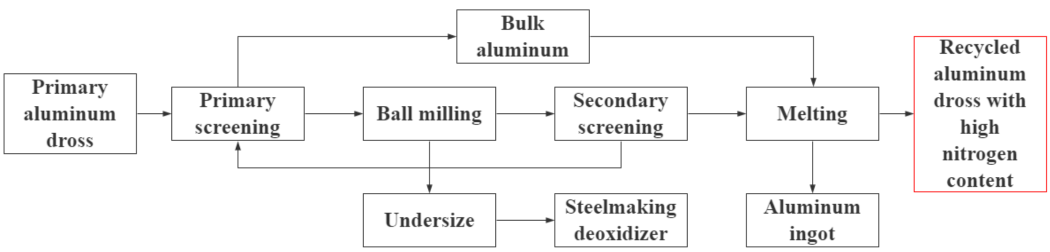

Figure 1. Production flow chart of aluminum dross.

Reagent: sodium carbonate (analytical grade, Jiangsu Qiangsheng Functional Chemical Co., Ltd., Suzhou, China), calcium oxide (analytical grade, Tianjin Damao Chemical Reagent Factory, Tianjin, China), sodium hydroxide (analytical grade, Xilong Chemical Co., Ltd. Shantou, China), hydrochloric acid (analytical grade, Shanghai Lingfeng Chemical Reagent Co., Ltd., Shanghai, China), methyl red (indicator, Shanghai Yuanye Biotechnology Co., Ltd., Shanghai, China), methylene blue (indicator, COOLABER SCIENCE&TECHNOLOGY) and boric acid (chemical pure, Shanghai Merrill Chemical Technology Co., Ltd., Shanghai, China).

Instrument: Program-controlled batch-type furnace (SXL-1230, Hangzhou Zhuochi Instrument Co., Ltd., Hangzhou, China), universal electric furnace (DK-98-II, Jiangsu Xinghua Chushui Electric Thermal Appliance Factory, Taizhou, China), ultrapure water machine (EPED-10TH, Nanjing Yipu Yida Technology Development Co., Ltd., Nanjing, China), X-ray fluorescence spectrometer (ZSX PRIMUS III+, Rigaku Electric Co., Ltd., Beijing, China), X-ray diffractometer (D8 Advance, Bruker Technology Co., Ltd., Beijing, China) and field emission scanning electron microscope system (ZEISS sigma HD, Carl Zeiss Management Co., Ltd., Shanghai, China).

XRD: the maximum output power is 3 kW, the voltage is 20–60 kV, the current is 2–60 mA, and the radius of the goniometer is 185 mm. MDI jade 9 was used for XRD analysis. XRD database is PDF 2009.

SEM-EDS: the acceleration voltage is 10 kV, the magnification is 100–200 k, the secondary electron resolution is 1 nm.

2.2. Experimental Method

The experiment mainly studies the effects of mixture ratio, roasting temperature and roasting time process parameters on the roasting denitrification effect of aluminum dross. The specific experimental steps are as follows:

(1) The additive and fully dried aluminum dross are ground according to the mixture ratio ($m_{additive}$:$m_{aluminum\ dross}$). 50 g homogenized mixture is extracted and placed in the corundum crucible.
(2) The crucible is placed in a batch-type furnace for firing at a certain temperature.
(3) The crucible is removed from the batch-type furnace after firing for a certain period of time and then cooled to room temperature in air.
(4) The cooled aluminum dross is ground into small pieces to complete the calcined sample. The samples are stored in a desiccator for subsequent detection of the denitrification rate, microscopic morphology and material composition of aluminum dross.

2.3. Detection Method

The acid-base constant volume method is used to determine the denitrification rate of aluminum dross. The specific operations are as follows [18]:

(1) First, 2 g aluminum dross is weighed out by balance and then poured into a conical flask containing 150 mL 20% NaOH solution. The stopper needs to be capped tightly.
(2) The conical flask is placed on a universal electric stove and heated until the solution boils. The solution is kept boiling for 2 h for distillation. The distilled ammonia gas is absorbed with 200 mL 40 g/L boric acid solution.
(3) After distillation, 0.05 mol/L dilute hydrochloric acid solution is used for titration, and standard methyl red-methylene blue is used as an indicator. During the titration, the endpoint of the titration is that the solution changes from blue to purple.

The calculation of AlN content and denitrification rate in aluminum dross is as follows:

$$W_1 = \frac{2.05C(V_2 - V_1)}{bM} \times 100\% \quad (1)$$

$$X_b = \frac{W_2 - W_1}{W_2} \times 100\% \quad (2)$$

where, W_1 is the aluminum nitride content in the sample (100%); V_1 is the volume of dilute hydrochloric acid consumed by the test sample (mL); V_2 is the volume of dilute hydrochloric acid consumed by the blank control group (mL); C is the concentration of dilute hydrochloric acid (mol/L); M is the mass of the test sample taken during measurement (g); b is the total mass of aluminum dross added to the test sample under the current mixture ratio (g); X_b is the denitrification rate (100%); W_2 is the aluminum nitride content (100%) in the original aluminum dross.

3. Results and Discussion

3.1. Organization and Composition of Aluminum Dross

XRD, XRF and SEM-EDS were used to analyze aluminum dross. The XRF test results are shown in Table 1, and the XRD test results are shown in Figure 2. It can be seen from Table 1 and Figure 2 that the composition of aluminum dross is relatively complex, mainly containing Al, Si, Cl and other elements, of which the content of Al element accounts for 61.69% at most, and the main phases are Al_2O_3, AlN and NaCl, while the single substance AI phase is relatively small, indicating that the AI element in aluminum dross mostly exists in aluminum-containing compounds, not in the form of an aluminum single substance, so the regenerated aluminum dross is not suitable for remelting and recovering metal aluminum. The SEM-EDS detection results are shown in Figure 3. Aluminum dross is composed of relatively independent particles of different sizes and shapes. The spherical particles in the red frame area in Figure 3a are mainly composed of elements Al, O and N. SEM, micro-area element surface distribution and EDS energy spectrum analysis are shown in Figure 3b,c, respectively.

Table 1. XRF analysis results of original aluminum dross (Unit: wt%).

Element	Al	Si	Cl	Na	Mg	Ca	Fe	K	Ti	Other
Content	61.69	8.58	7.70	4.30	3.32	3.13	2.77	2.47	1.26	4.78

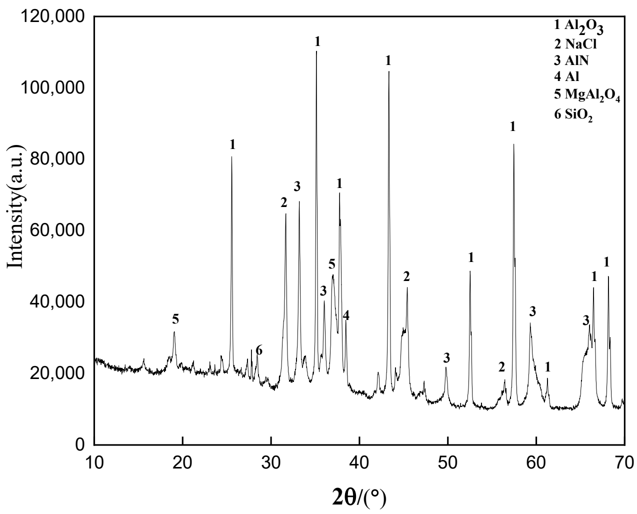

Figure 2. XRD pattern of original aluminum dross.

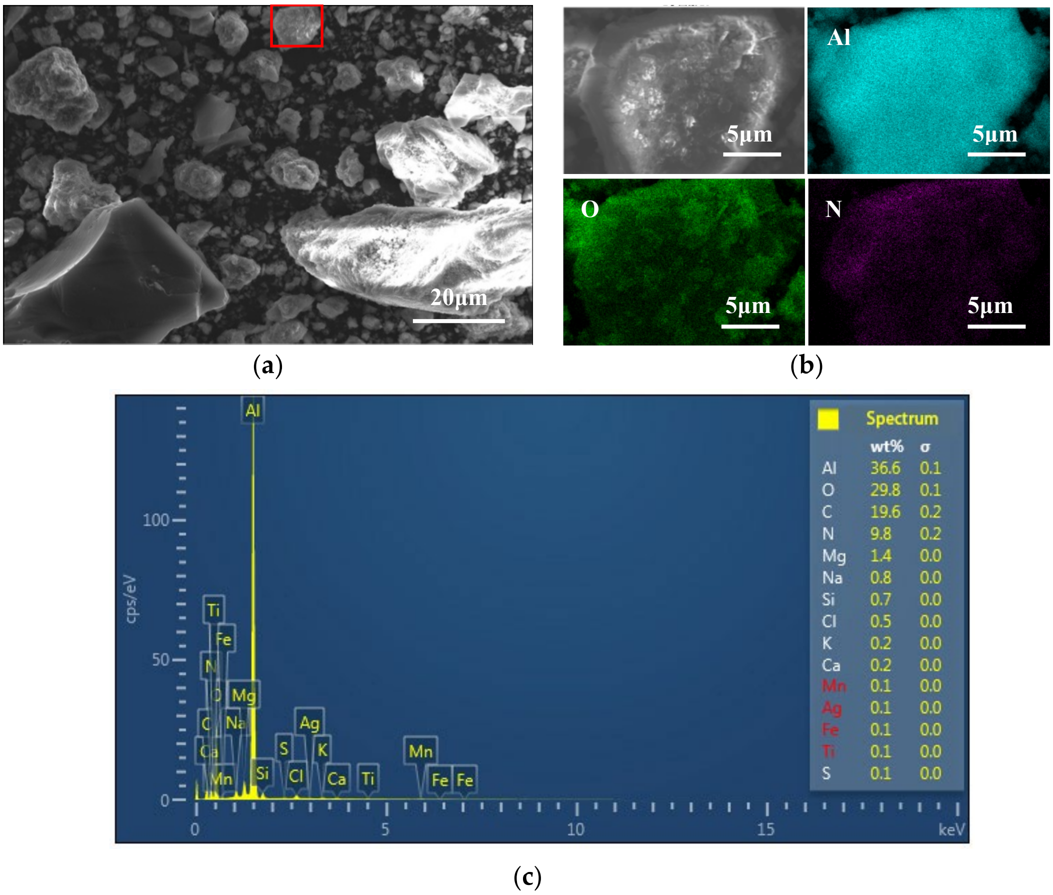

Figure 3. SEM-EDS image of original aluminum dross. (**a**) SEM of original aluminum dross; (**b**) SEM and micro-area element surface distribution of the red frame area; (**c**) EDS spectrum analysis of the red frame area.

SEM-EDS scanning results combined with XRD detection results show that Al, O and n elements in aluminum dross are mixed and doped with each other. AlN and Al_2O_3 in aluminum dross do not exist alone but are mixed with each other to form a complex mixture. Due to the relatively stable performance of Al_2O_3, the direct roasting and denitrification effect of aluminum dross is poor when Al_2O_3 wraps AlN. Therefore, additives need to be added to break the shell wrapped in AIN to improve the roasting and denitrification efficiency.

The acid-base constant volume method is used to determine the AlN content in the original recycled aluminum dross. With alumina as the blank control group, the results are shown in Table 2.

Table 2. Test results of AlN content in original recycled aluminum dross.

	Raw Material Aluminum Dross	Alumina
Hydrochloric acid consumption/mL	149.6 148.1 148.6	13.4 13.0 12.9
Average hydrochloric acid consumption/mL	148.76	13.1
AlN content/%	13.9%	/

3.2. Effect of Sodium Carbonate and Calcium Oxide on Roasting Denitrification of Aluminum Dross with High Nitrogen Concentration

3.2.1. Effect of Mixture Ratio on Denitrification of Aluminum Dross

The mixture ratio is the most important parameter used as an additive to speed up the denitrification rate of aluminum dross. An excessive mixture ratio will cause the additive to fail to react with the substances in the aluminum dross, resulting in a waste of resources. Too small a mixture ratio will result in poor nitrogen removal. Under constant roasting temperature and roasting time, the effect of the change of additive mass ratio to aluminum dross on the denitrification rate of aluminum dross by the mixture ratio is studied. Aluminum dross with a mixture ratio of 0.2, 0.4, 0.6, 0.8 or 1 is calcined at 900 °C for 4 h. The variation of the denitrification rate of aluminum dross with different mixture ratios is shown in Figure 4.

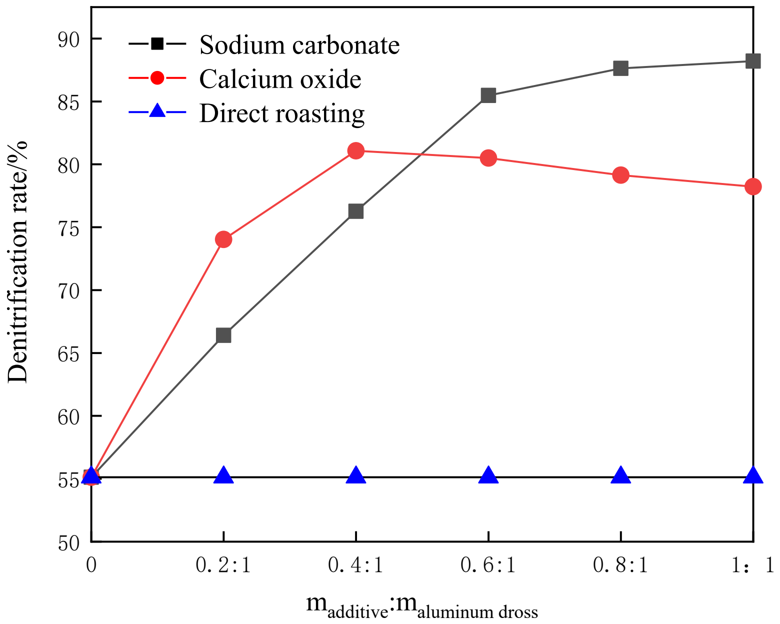

Figure 4. Denitrification rate of aluminum dross varies with mixture ratio.

It can be seen from Figure 4 that the denitrification rate of aluminum dross increases with the increase of the added amount of sodium carbonate. When $m_{sodium\ carbonate}$:$m_{aluminum\ dross}$ = 0.6, the denitrification rate of aluminum dross reaches 85.48%. The denitrification rate of aluminum dross does not increase significantly when sodium carbonate continues to increase. Under the same process conditions, the denitrification rate of direct roasting aluminum dross is only 55.12%. Na_2CO_3 can chemically react with Al_2O_3 at a certain temperature, as shown in formula (3).

$$Al_2O_3 + Na_2CO_3 = 2NaAlO_2 + CO_2 \quad (3)$$

Thermodynamic software HSC6.0 is used for the calculation of formula (3). Na_2CO_3 and Al_2O_3 can react spontaneously to form $NaAlO_2$ at 800 °C. The generated $NaAlO_2$ is an important solid phase for dissolving Al_2O_3, the Al_2O_3 shell wrapped with AlN can be broken open and more AlN can be directly exposed to the air, so the denitrification rate is improved [19]. Therefore, $m_{sodium\ carbonate}$:$m_{aluminum\ dross}$ = 0.6 is the optimal mixture ratio for roasting denitrification with sodium carbonate added.

For the samples added with calcium oxide, the denitrification rate of aluminum dross increases first and then decreases with the increase in the mixture ratio. At the same time, through the experimental phenomenon, it is observed that the agglomeration hardening degree of the treated sample is opposite to the change in denitrification rate, which first decreases and then increases. When the proportioning ratio was 0.2, the denitrification rate was 74.03%. When the blending ratio was 0.4, the agglomeration and hardening degree of the sample was the lowest, and the maximum denitrification rate was 81.08%. The ratio of ingredients continued to increase, and the denitrification rate began to decrease. When the proportioning ratio was 0.6 and 0.8, the denitrification rates were 80.5% and 79.14% respectively. Considering that CaO cannot react directly with AlN, the above phenomena show that the denitrification rate of aluminum dross is affected by the content of metal aluminum and calcium oxide in the material.

The oxidation rate of AlN in aluminum dross can be affected by the environmental atmosphere. The more fully AlN contacts with O_2, the faster its oxidation rate. When the mixture quality is constant, the greater the batching ratio, the lower the proportion of metal aluminum in the mixture, and the smaller the adhesion and wrapping of metal aluminum to the flowing aluminum dross particles after melting under this condition, so as to increase the contact opportunity between AlN in aluminum dross and O_2 in the air and improve the denitrification rate. Continuing to increase the mixture ratio leads to an increase in the proportion of CaO in the mixture. At this time, the particle size of calcium oxide powder is significantly lower than that of aluminum dross. Small calcium oxide powder fills the gaps in the aluminum dross, causing the material to become denser. The reduced AlN contact with O_2 reduces the denitrification rate [20]. Therefore, the optimal mixture ratio of aluminum dross treated with calcium oxide is $m_{calcium\ oxide}$:$m_{aluminum\ dross}$ = 0.4.

To sum up, both Na_2CO_3 and CaO can speed up the roasting denitrification rate of aluminum dross. Among them, Na_2CO_3 reacts with Al_2O_3, so that more AlN is exposed to the air, which improves the denitrification rate. And CaO is to reduce the degree of encapsulation of aluminum dross particles after melting by changing the metal aluminum content in the mixture, thereby improving the denitrification rate. In contrast, adding Na_2CO_3 denitrification is more direct and the effect is better.

3.2.2. Effect of Roasting Temperature on Denitrification of Aluminum Dross

In the case of roasting time = 4 h and additive ratio = 0.6(sodium carbonate)/0.4(calcium oxide), the temperature is set to 600, 700, 800, 900 and 1000 °C to study the denitrification effect of aluminum dross by roasting temperature, as shown in Figure 5.

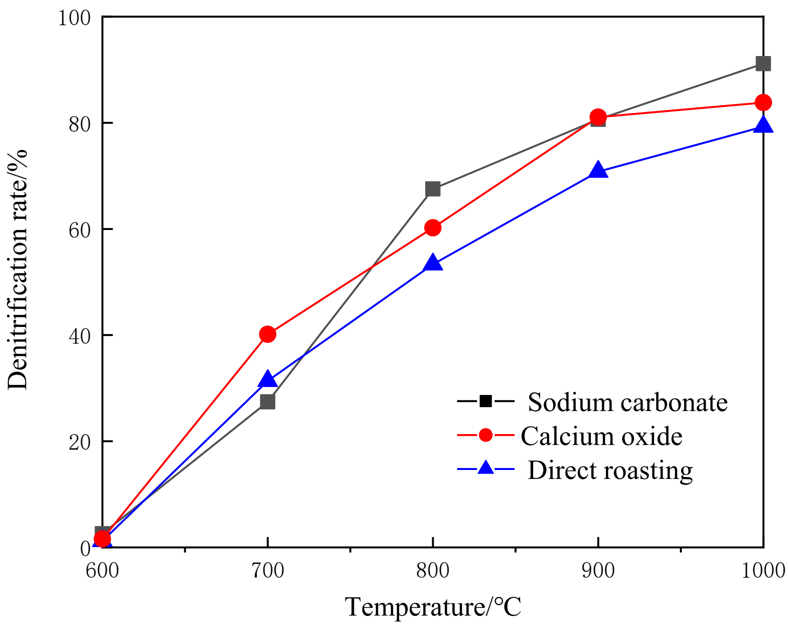

Figure 5. Denitrification rate of aluminum dross varies with roasting temperature.

According to Figure 5, the denitrification rate of the calcined samples with added sodium carbonate gradually increased with increasing temperature. According to the thermodynamic analysis, when the temperature is low, sodium carbonate and Al_2O_3 cannot spontaneously produce stress and do not damage the dense oxide film of Al_2O_3. Therefore, the denitrification effect is similar to that of direct roasting denitrification without substantial change. When the temperature exceeds 700 °C, sodium carbonate and Al_2O_3 react, the dense Al_2O_3 oxide film wrapped in AlN is broken, and the denitrification rate increases sharply. At this time, the denitrification effect is better than that of direct roasting. When the temperature rises to 1000 °C, the denitrification rate can reach 91.15%. In contrast, the direct roasting denitrification rate at this time is only 79.28%. Therefore, the optimal roasting temperature is 1000 °C.

For calcium oxide-added samples, the denitrification rate of aluminum dross increases with temperature. At 300 °C, the denitrification rate of aluminum dross is only 0.98%, which shows that AlN in aluminum dross oxidizes slowly under this condition. At 500 °C, the denitrification rate of the sample is not much different from that of direct roasting, with a denitrification rate of 1.59%. At 700 °C, the metal aluminum reaches the melting point and begins to melt under this condition. After melting, the degree of adhesion and wrapping of the aluminum dross particles decreases, thereby increasing the contact opportunity between AlN in the aluminum dross and O_2 in the air. At this time, the denitrification rate begins to appear significantly. increased, reaching 40.14%. At 900 °C, the difference between the denitrification rate (81.03%) of the roasted samples and that of direct roasting with calcium oxide addition is 10.24% at most. When the temperature continues to increase, the denitrification rate increases significantly slower [21]. Considering the effect of nitrogen removal and processing cost, the optimal roasting temperature is 900 °C. To sum up, the roasting temperature has a significant effect on the roasting denitrification rate of aluminum dross. The effect of both additives in the low-temperature stage was not obvious. As the temperature gradually increases the denitrification rate increases sharply. Through the analysis of the test results, the denitrification effect of Na_2CO_3 is far better than the denitrification effect of direct roasting and slightly better than that of CaO.

3.2.3. Effect of Roasting Time on Denitrification of Aluminum Dross

When the mixture ratio = 0.6 (sodium carbonate) and the roasting temperature = 1000 °C/ mixture ratio = 0.4 (calcium oxide) and the roasting temperature = 900 °C, the roasting time is set as 1 h, 2 h, 3 h, 4 h or 5 h to study the effect of roasting time on the denitrification of aluminum dross, as shown in Figure 6.

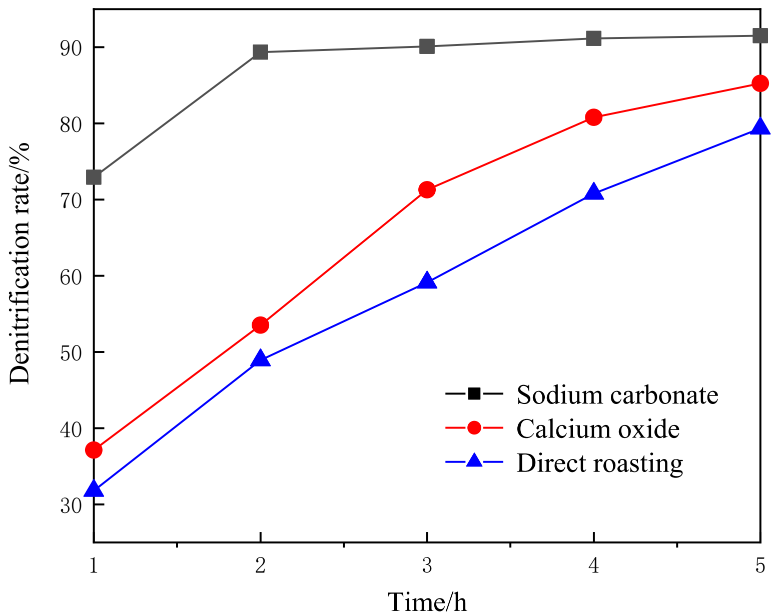

Figure 6. Denitrification rate of aluminum dross as a function of roasting time.

It can be seen from Figure 6 that at the roasting temperature of 1000 °C, sodium carbonate can accelerate the roasting denitrification of aluminum dross. With increasing temperature, the denitrification rate of both samples roasted with sodium carbonate and direct roasting increases. When roasting for 1 h, the denitrification rate of the sample added with sodium carbonate reaches more than 70%, while the sample of direct roasting is only about 30%, with an obvious gap. After calcination for 2 h, the denitrification rate of the sample added with sodium carbonate does not change significantly, which is stable at around 90%. At this time, its denitrification effect is still better than that of direct roasting. The treated samples are found to have increased whiteness with increasing roasting time. Considering that aluminum dross is black, it can be concluded that the reason for this phenomenon is the volatilization of certain impurities in aluminum dross. Therefore, the optimal denitrification process parameters of sodium carbonate-added aluminum dross are: $m_{sodium\ carbonate}:m_{aluminum\ dross}$ = 0.6, roasting temperature = 1000 °C and roasting time = 3 h.

For calcium oxide-added samples, the denitrification rate of aluminum dross increases with time, with a gradually slowing rate. When the roasting time is 1, 2 and 3 h, the denitrification rates of aluminum dross are 37.12%, 53.53% and 71.29%, respectively. When the roasting time is 5 h, the denitrification rate of aluminum dross reaches the highest (85.25%). Therefore, the optimal denitrification process parameters of calcium oxide-added aluminum dross are: $m_{calcium\ oxide}:m_{aluminum\ dross}$ = 0.4, *roasting temperature* = 900 °C and *roasting time* = 5 h.

3.3. Microstructure and Composition of Calcined Samples

Under $m_{sodium\ carbonate}:m_{aluminum\ dross}$ = 0.6, *roasting temperature* = 1000 °C and *roasting time* = 3 h, XRD detection of the roasted samples with sodium carbonate added is carried out, as shown in Figure 7a. The roasted samples with calcium oxide added under $m_{calcium\ oxide}:m_{aluminum\ dross}$ = 0.4, *roasting temperature* = 900 °C, *roasting time* = 5 h are taken for XRD detection, as shown in Figure 7b.

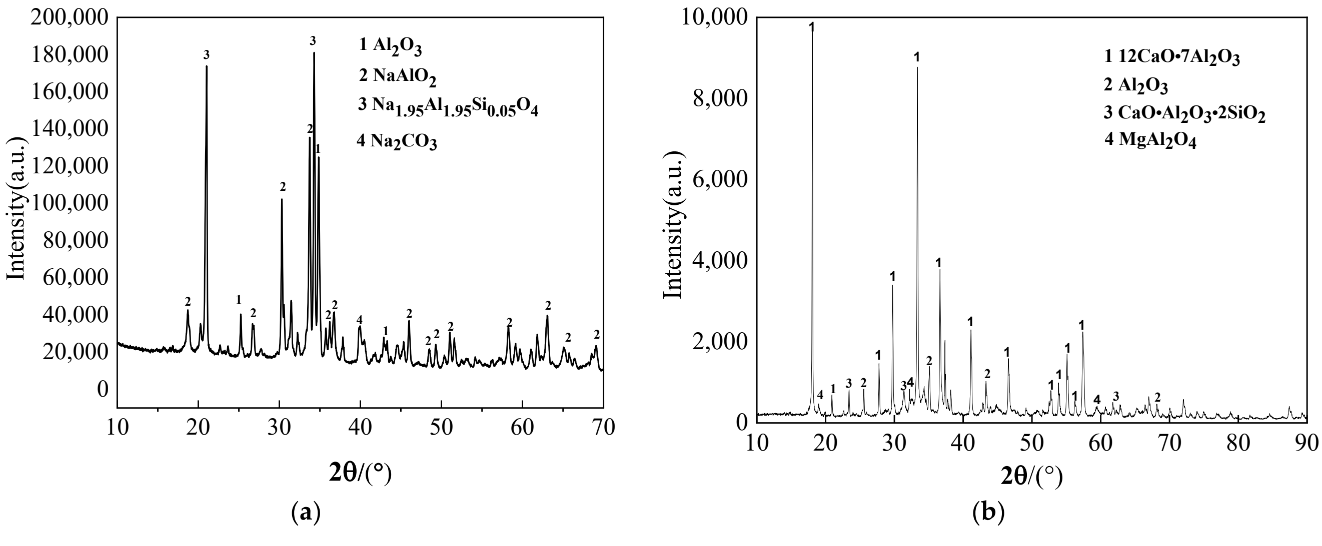

Figure 7. XRD of roasted samples with sodium carbonate and calcium oxide. (**a**) Add sodium carbonate; (**b**) Add calcium oxide.

No Al, AlN and NaCl phases were found in the samples, indicating that under this condition, Al in the aluminum dross slag had all reacted with O_2 and N_2 to form AI_2O_3 and AIN, see reaction formulas (4) and (5). AlN is oxidized to Al_2O_3 and shows the disappearance of the phase. The disappearance of the NaCl phase is the reduction of the volatilization of the substance at high temperature, which corresponds to the observed increase in the whiteness of the sample, indicating that the high-temperature waste gas generated during the roasting process not only contains N_2 and various nitrogen oxides but also contains a variety of aluminum dross slag. Due to the impurities volatilized during roasting, the waste gas needs to be treated during the treatment process to prevent environmental pollution.

$$4Al + 3O_2 = 2Al_2O_3 \quad (4)$$

$$2Al + N_2 = 2AlN \quad (5)$$

For the new phases $NaAlO_2$, $Na_{1.95}Al_{1.95}Si_{0.05}O_4$, $12CaO{\cdot}7Al_2O_3$ and $CaO{\cdot}Al_2O_3{\cdot}2SiO_2$, the samples added with sodium carbonate and calcium oxide were further analyzed by SEM-EDS.

SEM-EDS analysis of the sodium carbonate-added sample is shown in Figure 8. The main elements contained in the scanning area are O, Al, Na and Si, of which O content is the highest (52.26%), followed by Al and Na (13.62% and 13.41%) and Si (2.36%). According to Figure 7a, $NaAlO_2$ and $Na_{1.95}Al_{1.95}Si_{0.05}O_4$ are mixed together. Through the analysis of EDS scanning results, the Si content is much lower than the Al content. Therefore, the formation of $NaAlO_2$ from sodium carbonate and alumina is the main reaction during calcination. SiO_2 react with sodium carbonate and alumina to generate $Na_{1.95}Al_{1.95}Si_{0.05}O_4$, see reaction formula (6) [22].

$$1.95NaCO_3 + 1.95Al_2O_3 + SiO_2 = 2Na_{1.95}Al_{1.95}Si_{0.05}O_4 + 1.95CO_2 \quad (6)$$

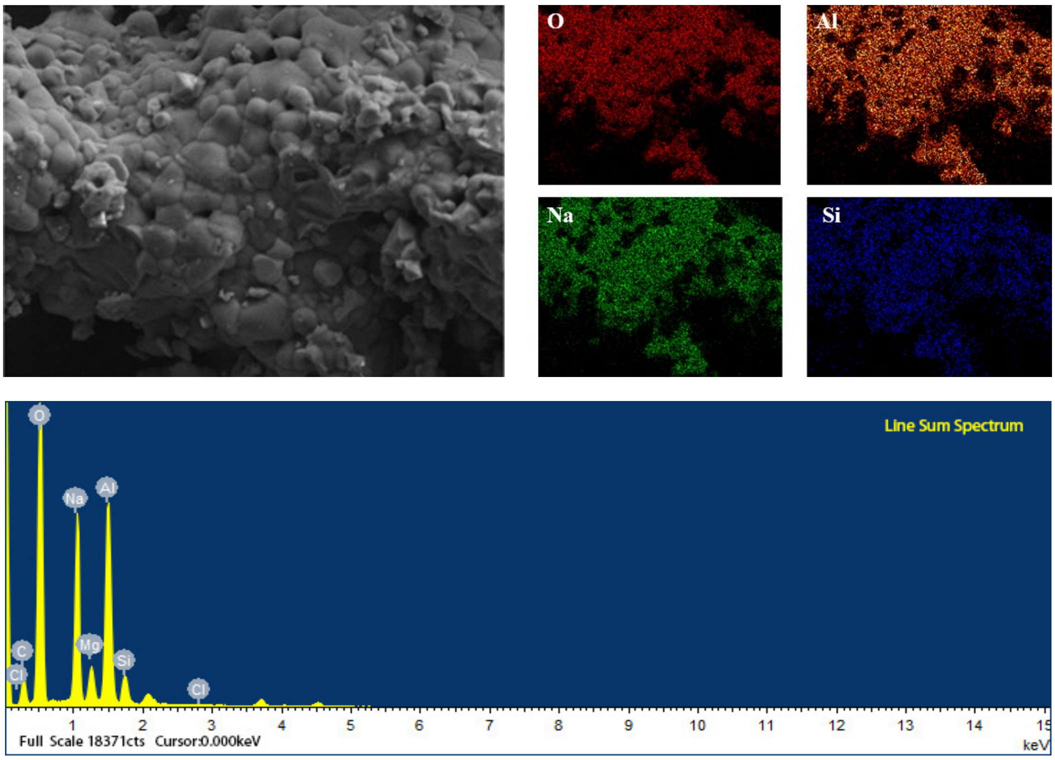

Figure 8. SEM-EDS image of the roasted sample with sodium carbonate added at $m_{sodium\ carbonate}:m_{aluminum\ dross} = 0.6$, *roasting temperature* = 1000 °C, *roasting time* = 3 h.

SEM-EDS analysis of the calcium oxide-added sample is shown in Figure 9. The aluminum dross particles appeared fine and aggregated after adding calcium oxide and calcined. The red frame area in Figure 9 is further subjected to SEM and micro-area element surface distribution, as well as EDS energy spectrum detection. According to the results, the main elements contained in the red box area are Al, O and Ca. Combined with the analysis of Figure 7b, it is believed that the addition of calcium oxide calcination can convert the difficult leaching Al_2O_3 in the aluminum dross into easily leaching $12CaO{\cdot}7Al_2O_3$, and at the same time can generate by-product $CaO{\cdot}Al_2O_3{\cdot}2SiO_2$, as shown in the reaction formula (7) and (8).

$$12CaO + 7Al_2O_3 = 12CaO{\cdot}7Al_2O_3 \quad (7)$$

$$CaO + Al_2O_3 + 2SiO_2 = CaO{\cdot}Al_2O_3{\cdot}2SiO_2 \quad (8)$$

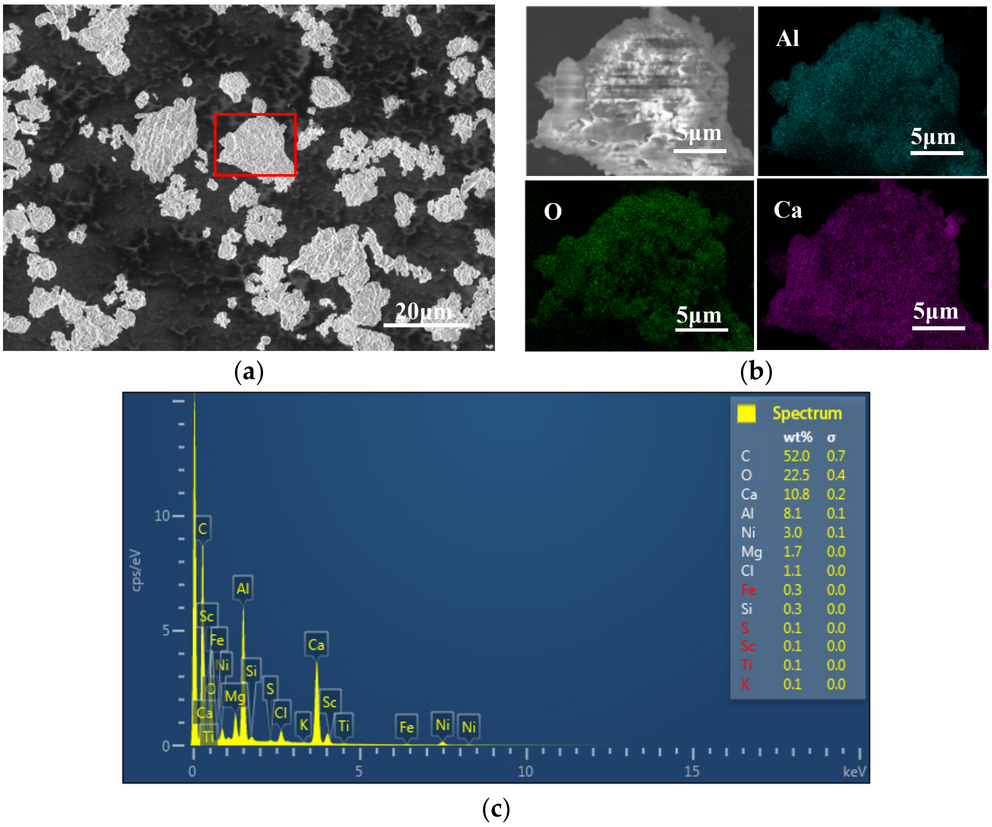

Figure 9. (**a**–**c**) SEM-EDS image of calcium oxide-added roasted sample under $m_{calcium\ oxide}$:$m_{aluminum\ dross}$ = 0.4, *roasting temperature* = 900 °C, *roasting time* = 5 h.

4. Conclusions

(1) The addition of sodium carbonate and calcium oxide can speed up the roasting denitrification rate of aluminum dross, which can be converted into valuable substances such as $NaAlO_2$ and $12CaO{\cdot}7Al_2O_3$ after roasting. In the process of treatment, both methods will produce high-temperature exhaust gas containing a variety of nitrogen oxides and impurities in aluminum dross. Therefore, it is necessary to collect and treat the gas in the treatment process to prevent environmental pollution.

(2) Adding sodium carbonate can directly destroy the oxide film wrapped on the surface of AlN, improve the contact area between ain and air, and improve the denitrification rate of aluminum dross.

(3) Calcium oxide mainly depends on reducing the wrapping degree of molten aluminum in the sample to make AlN more fully in contact with O_2 in the air, thus increasing the denitrification rate of aluminum dross. It further shows that the lower the content of metal aluminum in aluminum dross, the better the denitrification effect of aluminum dross treated by the calcium oxide roasting method and the lower the roasting temperature and less energy consumption.

(4) The optimal denitrification process parameters of the two additives are obtained by the single factor optimization experiment method. Under $m_{sodium\ carbonate}$:$m_{aluminum\ dross}$ = 0.6, *roasting temperature* = 1000 °C and *roasting time* = 4 h, the denitrification rate can

reach 91.32%. Under $m_{calcium\ oxide}$:$m_{aluminum\ dross}$ = 0.4, *roasting temperature* = 900 °C and *roasting time* = 5 h, the denitrification rate can reach 85.25%.

(5) The research results have certain guiding significance for aluminum dross harmless treatment and have reference value for the research and development of new aluminum dross harmless treatment equipment and resource utilization equipment.

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