



Article Cementitious Behavior of Argon Oxygen Decarburization Stainless Steel Slag and Its Stabilization on Chromium

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Abstract: The study mainly aims at the potential of Argon Oxygen Decarburization Slag (AODS) as a supplementary cementitious material and explores the mechanisms of stabilization/solidification (S/S) of chromium in cement-based composite pastes. The basic cementitious parameters, such as water requirement, setting time, soundness, hydration characteristics, and strength indexes of composite binders, were examined through standard methods. The results showed that the most beneficial mineral phase in AODS for cementitious behavior was beta dicalcium silicate (β -C₂S). The utilization of a higher AODS dosage in composite binders increased the water requirement and the setting time, while it decreased the hydration heat and the strength indexes. Although the AODS possessed limited cementitious properties, it conformed the Grade II steel slag powder qualified for concrete and cement. Sequential leaching tests were conducted targeting the leachability of chromium in the pastes with different AODS dosage and curing time. Results showed that with the lower AODS dosage and the longer curing time, the S/S efficiency for chromium leaching from the composite paste was better. Utilization of AODS as a cement substitute not only can recycle this solid waste and decrease the emission of CO₂ concerning cement production, but also helps to effectively reduce the chromium leaching risk.

Keywords: AOD slag; cementitious property; stabilization/solidification; chromium; leachability

1. Introduction

Stainless-steel slag includes Electric Arc Furnace Slag (EAFS), Argon Oxygen Decarburization Slag (AODS), and Ladle Metallurgy Slag (LMS) [1]. AODS is generated in an AOD furnace in the refining process of stainless-steel. It has a high content of CaO, SiO₂, and MgO, as well as some Cr₂O₃ and fluorine [2]. In its cold state, the crystalline mineral phase composition of AODS is mainly γ -dicalcium silicate (γ -C₂S), β -dicalcium silicate (β -C₂S), periclase and fluorite, traces of f-CaO, merwinite, and spinel [3]. In the cooling process, C₂S transforms from β -type to γ -type around 490 °C [4]. This mineral phase transformation causes an increase of about 12% in volume, causing shattering of the crystals and generating the powdery AODS [5]. The quantity of AODS produced in China annually accounts for about 1.8 million tons, containing an approximate amount of 9540-ton chromium (Cr) [6]. Once the Cr enters the environmental cycle, trivalent Cr can be oxidized to hexavalent Cr with oxygen [7]. Hexavalent Cr has been identified as a toxicant that can induce carcinogenic and toxic impacts through a complex multiple mechanism of action including mutagenesis, oxidative stress, damage on chromosome and DNA, and epigenetic changes [8].

The physicochemical properties of alternative materials determine their potential as cementitious additives in cement-based binders [9,10]. According to the physical and chemical characteristics of AODS, it can be classified as a kind of industrial by-product that can be used in cement-based applications [11]. The types and contents of oxides in AODS are like Portland cement (PC). Due to the presence of hydratable C₂S phases, AODS has cementitious properties [12]. In previous research, the specific surface area of fresh AODS was determined to be 511 m²/g [13], which facilitates its application as a cement substitute [14].

It is vital to observe that γ -C₂S is the major C₂S form in the AODS, accounting for approximately 38% of the slag mass [15]. In general, γ -C₂S is difficult to hydrate, and its hydration ratio in aqueous media for five years is only 20–25% [16]. β -C₂S is hydraulic, but its content in AODS is much lower than that of γ -C₂S [17]. Therefore, the application potential of γ -C₂S in cement-based materials directly determines the applicability of AODS. γ -C₂S can be easily carbonated without hydration when in contact with CO₂ and water [18]. Using carbonation properties, many researchers have found that γ -C₂S can be used as a durability modifier for a cement-based product. Mabudo et al. [19] researched the properties and structure of cement-based mortars and pastes with different γ -C₂S supplementary ratios under atmospheric carbonation curing and found that carbonation curing reduced porosity by about 33% and improved the mechanical properties of composite materials from 50 MPa to 70 MPa. For cement-based products with lower initial strength but higher durability requirements, it may be a good choice to use AODS as a cementitious substitute.

The significance of applying such industrial waste as a cement substitute is that it can save landfill space [20,21] and decrease CO_2 emissions in the cement production [22–24]. Except for cementitious characteristics, the potential environmental issues related to the leaching risk of chromium must be considered when AODS was utilized as a cement substitute. The recycling of AODS was very limited due to its high Cr content (0.5–5 wt.%) [13,25–27]. Studies have shown that the chromium leaching risk of AODS is usually underestimated [7]. Under certain leaching conditions, the concentration of chromium in the leachates obtained from the AODS products is enough to pose an ecological risk [28].

The stabilization/solidification (S/S) process is one of the effective technologies to treat industrial wastes containing hazardous materials [29–33]. The S/S process plays its role by mixing solid waste with cementitious materials to reduce mobility, as well as the toxicity of the contaminants [32,34], and this process usually accompanies encapsulation, absorption, precipitation, and finally detoxification [35,36]. S/S by cementitious materials like cement is in an extensive application for immobilization of chromium [37,38]. Chromium can be efficiently immobilized by the hydration products of the cured cement-based composite cementitious materials [39]. Applying the AODS as the partial substitute for cement is an S/S process. The existence of chromium in AODS has its characteristics [40], but researchers also need to obtain information on how chromium is present in the cement-based composite material that has undergone the S/S process to beneficial assessing chromium leachability. While studying the maximum applicability of AODS in cement, it is also necessary to consider the potential ecological risks of chromium introduced by the incorporated AODS [26].

The specific focus of this research is on evaluating the cementitious property of AODS and analyzing the S/S effect on the chromium sequestration in the cement-based composite binders with different AODS dosages. The water requirement, setting time, soundness, and activity indexes of composite binders were tested by using a series of standard test approaches. The hydration heat evolution of composite binders was monitored adopting a TAM-Air isothermal calorimeter. Changes of mineral phases and their microstructures of the hydrated pastes were analyzed through Fourier-transform infrared (FT-IR) spectroscopy, scanning electron microscopy (SEM/EDS), and X-ray diffraction (XRD). The hydration ratios of the pastes were determined by thermogravimetric (TG/DTG) analysis. Moreover, a sequential leaching test (EN-12457) was conducted to acquire information concerning Cr leachability in the pastes with various AODS supplemental and hydration ratios.

2. Experiment

2.1. Raw Materials

The material of AODS was directly collected in the stainless-steel refining workshop. The received AODS presented a powdery appearance with disaggregated particles. After being sieved by using a 120-mesh sieve, the AODS was continuously dried at a constant temperature of 105 °C for more than 5 h and later stored in a vacuum-sealed and dry environment.

The 42.5 strength grade Portland cement (PC), which conformed to the Chinese National Standard (CNS) GB175-2007, was used as the reference cementitious material. The chemical analysis of AODS and PC was carried out by using an Axios advanced X-ray fluorescence spectrometer (PANalytical B.V., Almelo, Netherlands), the result was quantified in oxide form. The free lime contents of AODS and PC were tested by the ethylene glycol method [41].

The distributions of particle size of AODS and PC were determined with a Mastersizer 2000 laser granularity meter (Malvern Panalytical, Malvern, UK). Absolute ethanol was used as the dispersion medium in the test.

The mineralogical compositions of AODS and PC were measured based on a combined method of XRD and FT-IR. The XRD test was performed on a D8 Advance X-ray diffractometer (Bruker Corporation, Billerica, Massachusetts, USA), and diffraction patterns were evaluated in the 2θ range of 10° to 90°. The crystalline phases were identified using JADE 9 software based on the ICDD Powder Diffraction File (PDF-2009) database. Fourier-transform infrared (FT-IR) measurements were performed on the composition of mineral phases, which played the role of supplementary verification. The vibration and rotational spectra of molecular in the samples were measured by using a Nexus FT-IR spectrometer (Thermo Nicolet, Madison, Wisconsin, USA) in the wave range of 4000–400 cm⁻¹.

2.2. Paste Samples

The reference binder (B0) was prepared from pure PC and five AODS dosages were adopted to prepare the test binders (B1, B2, B3, B4, and B5). Each dry binder was mechanically mixed in a blender for 30 min to reach complete homogeneity.

The normal consistency water requirement, the setting time, and the soundness of binders were tested in line with the CNS GB/T 1346-2011 (ISO 9597: 2008). The cementitious properties of AODS were obtained initially by analyzing the effects of AODS dosage on these three basic cementitious indexes. All of the analyses were performed in duplicate.

The hydration exotherm rates, together with the cumulative exotherms of the composite binders, were monitored through an isothermal calorimeter (TAM Air from TA Instruments) according to ASTM C1702 [42]. The ratio of water to binder was set to 0.4 mL/g and the measurements were implemented at the constant temperature environment of 20 °C within 96 h.

To systematically study the hydration products of the composite binder, the wet composite pastes were produced by mixing the composite binders and water with water to binder ratio of 0.4 mL/g. Then, the composite pastes were immediately sealed in plastic wraps. At test ages, absolute ethanol was used to immerse the hardened pastes to avoid further hydration before exsiccation of them in a vacuum drying oven at 50 °C for 48 h before further testing.

The measurement of hydrated pastes' mineral phases was conducted by XRD coupled with FT-IR, and the experimental parameters were described previously. The morphology and micromorphology of hardened pastes were scanned by the usage of a Quanta 650 FEG electron microscopy (Thermo Fisher, Waltham, MA, USA). The elemental compositions of representative products in hardened pastes were measured through energy-dispersive X-ray spectroscopy (EDS).

The hydration degree was determined by TG-DTG analysis. The analysis of TG was conducted by using a Setsys evo TG analyzer (Setaram, Lyon, France). Using a cylindrical Al2O3 crucible as a sample container, we heated the paste samples from ambient temperature to 950 °C protected by nitrogen atmosphere, and the heating rate was set to 0.33 °C/s.

2.3. Mortar Samples

The mortar was prepared by one part of the mixing binder (450 ± 2 g), three parts of ISO reference standard sand (1350 ± 5 g), and one-half part of deionized water (225 ± 1 g). After complete mixing, the resultant mortar was molded into three same shaped specimens by using a standard mold of 40 mm × 40 mm × 160 mm, which contained three same prismatic shape grooves. After being compacted on a jolting table, the molds containing the mortars were cured in a room with a temperature of 20 ± 1 °C and relative humidity higher than 95% for 24 h, then the hardened mortar specimens were de-molded and then cured underwater at 20 ± 1 °C for time intervals of 3, 7, and 28 days (including the 24 h for the setting). For parallel experiments, the binder of each AODS-PC formula was made into two sets of mortar specimens with the standard mold.

The reference mortar sample was prepared by using binder B0; other mortars with different AODS-PC dosages were prepared as test mortars. Compressive and flexural strength were tested upon mortar samples after curing for 3, 7, and 28 days. The activity indexes of these binders were calculated by Equation (1):

$$A = R_i / R_0 \times 100 \tag{1}$$

where A denotes the activity index of the AODS, R_i denotes the strength value of the test mortar at the corresponding curing age, while R_0 denotes the strength value of the reference mortar at the corresponding curing age.

2.4. Sequential Leaching

To study the effect of cementitious solidification on the leachability of chromium in composite binders, the sequential leaching tests designed according to European Standard EN12457-2 were conducted on pastes with different AODS dosages and hydration ratios.

2.4.1. Leaching Subjects

Hardened pastes prepared from B1, B2, B3, B4, and B5 curing for 28 days were dried at 105 $^{\circ}$ C for 6 h and then processed by crushing and grinding until the maximum particle size was <80 μ m. The obtained paste powders labeled as P1, P2, P3, P4, and P5 were used as the leaching subjects to study the effect of cementitious solidification on chromium leachability.

In addition to the dosage of the cementitious admixture, the hydration ratio—which was positively related to the curing time—was another important factor affecting the S/S on chromium leachability. Hardened pastes prepared from B5 after curing for 3, 7, and 28 days were chosen as the subjects to analyze the effect of the solidification process relating to hydration ratio on chromium leachability. The B5 dry powder without hydration was used as the reference leaching subject.

2.4.2. Leaching Procedure

Ten grams of hardened paste powder was weighed and placed into a 250-mL conical flask, with 100 mL deionized water added into the same flask. Then a lid was used to seal the flask to prevent the intrusion of oxygen and carbon dioxide. Then the sealed flask was fixed in a ZD-85 type oscillator, which can roller-table, inducing rotation of the conical flask at 10 rps. It was a 20-day sequential leaching test in a condition of room temperature. Every 24 h an 80 mL supernatant was aspirated for electrochemical and chromium concentration tests. To keep the L/S ratio at the same level in each sampling cycle, 80 mL of deionized water was supplemented accordingly every day.

After filtered, the leachate's electrochemical characteristics containing pH, redox potential (Eh), and electrical conductivity were examined. In addition, chromium concentration of the leachate was tested by inductively coupled plasma-optical emission spectrometry (ICP-OES) with a detection limit of 0.004 mg/L. All of the analyses were performed in duplicate.

3. Results and Discussion

3.1. Properties of Raw Materials

The chemical compositions of the AODS and PC are presented in Table 1. The main components in both AODS and PC were CaO and SiO₂, which made up more than 85% of the total mass. Higher SO₃ content in PC than AODS was due to the additional plaster provided in the clinker to slow down the early setting rate. The Cr_2O_3 content in the studied AODS was 0.5%, which was in the normal range [43]. The free lime content of AODS was 0.33%, even lower than that of PC. Thus, expansion due to hydration of the free lime was neglectable [44].

Table 1. Chemical composition of the Argon Oxygen Decarburization Slag (AODS) and Portland cement (PC) (wt. %).

Oxides	CaO	SiO ₂	MgO	Al ₂ O ₃	Fe ₂ O ₃	SO ₃	Cr ₂ O ₃	TiO ₂	f-CaO
AODS	65.78	24.15	6.55	1.74	0.26	0.54	0.50	0.22	0.33
PC	65.80	19.12	1.90	5.15	3.37	3.10	0.01	0.31	0.41

The particle size distributions of AODS and PC are shown in Figure 1. The particle size distribution interval was mainly between 0.5 μ m and 80 μ m. The AODS had more fine particles (<11 μ m) and fewer coarse particles (>11 μ m), and as a result, the mean particle size of the AODS was smaller than that of the PC. Specific surface areas reported by calculation software for AODS and PC were 511 m²/kg and 418 m²/kg, respectively.



Figure 1. The particle size distribution of AODS and PC.

The XRD patterns of AODS and PC are presented in Figure 2. It was found that dicalcium silicate was the predominant mineral phase in AODS, while fluorite, periclase, merwinite, and chromium spinel were also detected, supporting conclusions of other researches on AODS [3,13]. The four typical clinker mineral phases, including dicalcium silicate, tricalcium silicate, tricalcium aluminate, and calcium aluminoferrite, were all detected in the PC [45].

The FT-IR pattern of AODS is presented in Figure 3. By analyzing the FT-IR pattern, detailed information about the C₂S phase in the AODS was examined. The bonds around 927 cm⁻¹ and 816 cm⁻¹ corresponded to γ -C₂S, while the bond around 841 cm⁻¹ corresponded to β -C₂S [13].

On the basis of the results of the XRD and FT-IR analyses, the C₂S occurred in two allotropic forms (β -C₂S and γ -C₂S) in AODS. The content of the hydratable β -C₂S was much lower than the almost non-hydratable γ -C₂S.



Figure 2. X-ray diffraction (XRD) results of the AODS and PC.



Figure 3. Fourier-transform infrared (FT-IR) patterns of the AODS.

3.2. Cementitious Properties of AODS

3.2.1. Water Requirement, Soundness, and Setting Time

The water requirement, setting time, and soundness tests were performed on the composite pastes, and the results are presented in Table 2.

Table 2. Cementitious properties of binders with different formulas.

Binder	AODS (g)	PC (g)	Water Requirement (%)	Initial Setting Time (min)	Final Setting Time (min)	Expansion Ratio (%)	Soundness Determination
B0	0	450	27.5	144	216	0.35	Qualified
B1	50	400	29.11	163	231	0.30	Qualified
B2	100	350	30.52	181	246	0.34	Qualified
B3	150	300	31.73	190	265	0.38	Qualified
B4	200	250	32.54	198	282	0.41	Qualified
B5	250	200	33.06	203	301	0.43	Qualified

The water requirement contents of the composite pastes to reach standard consistency were positively correlated with the dosage of AODS. When the dosage of AODS reached 55.55%, the water requirement content increased from 27.5% of pure cement (B0) to 33.06% of B5.

The specific surface area of the PC was smaller than that of AODS. AODS particles required more surface-coated water than cement, which would lead to insufficient water content in the slurry to provide a lubrication function. Consequently, more water would be required for the paste to achieve the same consistency with the increase of AODS dosage.

The soundness of paste containing AODS is a critical parameter when it is used as the admixture for a binder. Soundness can be determined by the ability of the paste to maintain its volume after hardening. From the results presented in Table 2, it can be concluded that the AODS meets the soundness requirements (expansion ratios were <0.50%) specified by the CNS GB/T 750-92. This is mainly because of the free lime content of AODS only being 0.33%, meeting the standard requirement of less than 3%.

Table 2 shows that all the composite binders met the PC setting time specified in the CNS GB 175-2007. With the increasing of cement replacement ratio, the setting time was longer. This phenomenon was similar to the result reported in the previous literature [46]. AODS possess lower hydratable mineral content, but a larger specific surface area than PC. Therefore, as the incorporation ratio of AODS was increased, the total interface area between binder particles in the slurry state paste increased gradually. More C-S-H gel was required to produce more bonding functions at the interfaces to reach the setting state, but the added AODS cannot produce enough C-S-H gel to make up for this demand. Consequently, the setting time of the composite paste was prolonged by AODS admixture.

3.2.2. Hydration Exotherm

The hydration exothermic rate and cumulative hydration heat of the composite binders with different AODS dosages are presented in Figures 4 and 5, respectively.



Figure 4. Hydration exothermic rate of composite pastes within (a) 0-150 h, (b) 0-1 h, (c) 1-30 h, and (d) 30-150 h.



Figure 5. Cumulative hydration heat of (a) composite pastes and (b) selected referenced composite pastes.

The hydration process of pure AODS had only one exothermic peak. Owing to the greater specific surface area than cement, the AODS exothermic rate was much higher than that of PC throughout the rapid exothermic period (0–0.15 h) and the deceleration period (0.15–8 h). When the hydration of AODS entered its steady period (>8 h), its exothermic rate was negligible.

When the AODS dosage was lower than 33%, the hydration process of composite binders had a similar trend to pure cement. Two exothermic peaks could be observed in the hydration exothermic rate patterns of B1, B2, as well as B3. While when AODS dosage was higher than 44%, the hydration process of binders was more complicated, and three exothermic peaks could be observed for B4 and B5. The hydration steady period was continuously delayed as the AODS dosage increased, which accords with the results of the time setting analysis. For all composite binders, the exothermic rates decreased gradually during the deceleration period owing to the reduction in the surface area of the hydrating PC and AODS grains.

As shown in Figure 5a, the total heat release values for all composite binders were lower than that of pure PC, which was mainly related to differences of mineralogical composition between AODS and PC. The hydraulic capacity of the mineral phases in AODS, such as γ -C₂S, merwinite, Mg-Cr spinel, and fluorite, have been reported to be rather limited [28,42]. As the main hydratable mineral phase in AODS, β -C₂S's hydration efficiency was much lower than that of C₃S [47]. The cumulative hydration heat in 150 h decreased from 274 J/g for pure PC to 148 J/g for the binder B5.

The theoretical heat release values of the cement part in the composite binders were calculated using pure PC experimental data as a reference, and the comparison curves are shown in Figure 5b (take B1, B3, and B5 into account). The cumulative hydration released heat of composite binders was not as high as that of the cement part initially and then surpassed it. The intersection point would be delayed with the increase of AODS dosage. It can be concluded that the AODS had a certain hindrance to the hydration of the cement part in the early stage. According to studies of Zheng et al. [48] as well as Péra et al. [49], the mechanism of AODS substitutes effect on the retardation of cement hydration was related to its higher MgO content. When the hydration progressed to a certain degree, the cementitious minerals in the cement part enter an acceleration hydration period, plus the contribution of AODS hydration, and the cumulative hydration heat of composite binders surpassed that of the cement part. This indicated that the cementitious contribution of AODS was mainly manifest in the latter stage of hydration.

3.2.3. Hydration Products

Figure 6 illustrates the FT-IR spectrums of the composite B4's pastes with different curing ages. The three spectrums showed bands at 3640 cm^{-1} , 3435 cm^{-1} , 1638 cm^{-1} , 1440 cm^{-1} , 1422 cm^{-1} , 940 cm^{-1} , 875 cm^{-1} , and 500 cm^{-1} . The sharp bands that appeared around 3640 cm^{-1} were associated with the asymmetric stretching vibration of the OH⁻ groups in Ca(OH)₂ [50]. The broad bands near 3435 cm^{-1}

were related to the symmetric stretching water ([Al (OH₆)]) in the ettringite [51]. The bands appearing at 940 cm⁻¹ were attributed to the anti-symmetric stretching of the Si–O–Si bond in C-S-H gel. During the hydration curing procedure, the mineral phases with a cementitious ability, such as C₃A, C₃S, and β -C₂S, reacted with water to form calcium hydroxide, ettringite, and C-S-H gel. The band at 1638 cm⁻¹ corresponded to the bending vibration of the H–O–H band for the interlayer water [52]. It can be concluded that during the pastes' hydration curing, the moisture not only participated in the mineral hydration reaction, but also transformed into interlayer water, which cannot be removed completely at 105 °C.



Figure 6. FT-IR spectrums of the composite B4 pastes with different curing ages.

It should be noted that the spectral region 1422–1440 cm⁻¹ belonged to asymmetric stretching v_3 CO₃²⁻ from amorphous CaCO₃ [53], and the absorption peaks at 875 cm⁻¹ assigned to the out-of-plane bending of v_2 C–O from calcite and vaterite [54]. The curing environment of the composite pastes met all the requirements of mineral carbonation [13]. The carbonation phases in the composite pastes included the hydration products like Ca(OH)₂, unhydrated C₃S, unhydrated β-C₂S, and γ-C₂S. When these phases were in contact with the dissolved CO₂, the carbonation reaction occurred, and CaCO₃ was produced as one of the carbonation products. The bonds of the SiO₄⁴⁻ that appeared around 500 cm⁻¹ corresponded to the remaining silicate phases, such as β-C₂S, C₃S, and γ-C₂S [55,56].

The TG-DTG diagrams of the composite B4 paste differing in curing ages are shown in Figure 7. The DTG curves show that there were three major mass loss stages for the three aged samples with the increase of temperature. The first stage was in the temperature range of 55–300 °C, corresponding to the escape of interlayer moisture as well as chemical bonding water (ettringite and C-S-H gel) [57,58]. Because the aged paste samples had been dried at 105 °C for 6 h prior to TG-DTG analysis, the amount of interlayer water in the dried test paste samples was small. In consequence, the mass loss occurred at the initial step was mainly regarding the dehydration of the ettringite and C-S-H gel. The second mass loss stage detected in temperatures from 400 °C to 550 °C corresponded to the thermal decomposition of the Ca(OH)₂ [59]. It can be noted that the DTG peaks for Ca(OH)₂ were sharp, which indicated that the decomposition speed of Ca(OH)₂ was drastic. The third mass loss stage in the temperature range of 550–950 °C corresponded to the thermal decomposition of the Ca(OH)₂ [60,61].



Figure 7. Thermogravimetric (TG-DTG) analysis of composite B4 pastes with different curing periods.

3.2.4. Mechanical Strength of Mortars

To evaluate the specific strength of the mortars manufactured with the composite binders, compressive and flexural strength examinations were carried out (GB/T 17671-1999). Figure 8 indicates the results for the strength of mortars with different curing ages. The compressive and flexural strength was an increase with curing time and decrease with AODS dosage. The mortar prepared with B1 demonstrated a 28-day compressive strength of 42.51 MPa, which satisfies the requirements for complex Portland cement (P·C) 42.5 grade specified by GB175-2007. The composite binders B2 and B3 met the requirements for P·C 32.5 grade.



Figure 8. Flexural (a) and compressive (b) strength indexes of mortars with different AODS dosages.

The standard lines were obtained according to the PC dosages in the binders. At a curing age of 3 or 7 days, the activity indexes of the binders were not as high as that of the cement contents in the composite binders. This indicated that AODS harmed the mortar's strength in the early period. This was consistent with the results of hydration exotherm analysis. At a-28-day curing age, the activity indexes of the binders were close to, or even surpassed, the standard values. So, it can be concluded that mineral phases in the AODS hydrated and contributed to the strength of the mortars at late curing ages. In the early stage, the C_3S in the cement had undergone a hydration reaction to generate $Ca(OH)_2$, making the slurry alkaline. The alkaline environment had a certain activation effect on the AODS, which accelerated the hydration of the C_2S therein [42], and provided a favorable impact on the mortars' strength later in the process.

The steel slag (SS) powder ready to act as a substitute in cement can be categorized into two grades in accordance with the CNS GB/T 20491-2006. It specifies that the test binder sample is prepared by mixing 30% SS and 70% cement by mass, and activity indexes for the Grade I SS powder ought to be \geq 65% (curing ages of 7 days) and \geq 80% (curing ages of 28 days), separately. For the Grade II SS powder, activity indexes ought to \geq 55% (curing ages of 7 days) and \geq 65% (curing ages of 28 days), respectively. The 33.33% AODS-containing composite binder (B3) was used as the test sample to determine the cementitious grade, and the results showed that AODS could fulfill the requirements of the Grade II SS powder.

3.2.5. Morphology and Microstructure of Cured Pastes

The SEM photos of the B4's paste with different curing times are presented in Figure 9. According to SEM images, it could be noted clearly that gel-like structures, such as the C-S-H phase, had been formed. The paste hydrated with separated flakelike morphology at the early 3 days curing stage. As the hydration curing furthered to the 7th day, a reticulated network between the separated matrix was generated. As the hydration curing prolonged to the 28th day, the paste formed a denser agglomerated structure (see Figure 9c,d). In a word, as the curing period prolonged, the composite paste's porosity decreased gradually.



Figure 9. Scanning electron microscopy (SEM) photos of the B4's paste with a curing time of 3 days (**a**), 7 days (**b**), and 28 days (**c**,**d**).

Figure 10 shows the SEM images of 28-day-cured pastes with different AODS dosages. Combined with Figure 9d, it could be found that when AODS dosage was lower than 44%, the 28-day-cured paste appeared as a denser agglomerated structure with few pores. B5's 28-day-cured paste was ready to form a denser agglomerated structure, and the matrix tended to stack together. With the dosage of AODS increased to 55%, the needle-like C-S-H gel between the substrates was visible, and the reticulated networks far from enough to tightly connect the matrix. As the previous analysis shows, AODS had a much lower cementitious activity than PC, the paste with a larger AODS dosage cannot produce enough gelling substance to connect the matrix very well like the low AODS content pastes.



Figure 10. SEM photos of 28-days cured pastes with AODS dosage of 11% (a), 22% (b), 33% (c), and 55% (d).

3.3. The Chromium Leachability of Composite Pastes

3.3.1. Effects of Curing Time on Chromium Leachability

Figure 11 presents the evolution in pH, Eh, and electrical conductivity value of the leachates obtained from the 20-day sequential leaching test. As shown in Figure 11a, with the increasing of curing time, the leachate pH of the composite pastes increased, while their Eh decreased. Compared to the non-hydrated binder, the leachate pH of the 28-day-cured binder increased by ~0.5 pH units, which attributed to the continuous generation of Ca(OH)₂ during the curing procedure. More Ca(OH)₂ existed in the 28-day-cured paste, leading to a higher pH of the leachates. According to the study by [62], Eh is negatively correlated with pH, but positively correlated with the dissolved oxygen (DO) concentration. During the sequential leaching procedure, the increasing pH caused by the dissolution of Ca(OH)₂, coupled with the daily DO brought in during sampling and replacement of the leachates, increased the Eh of the leachates.



Figure 11. pH (**a**), Eh (**a**), and electrical conductivity (**b**) evolutions of the leachates for pastes with different curing time.

As shown in Figure 11b, the electrical conductivity, that could be used to measure the total ion leachability of composite pastes [63], sharply decreased from ~4.0 S/m to <0.5 S/m. This finding indicated a fact that numerous ions were rapidly released from composite pastes at the early leaching stage. As the leaching continued, a stage of sustained and slow release of ions was reached. Moreover,

it could be also found that electrical conductivity decreased with the increasing of the curing time. Furthered curing could generate enough hydration products, mainly C-S-H gel, to fill and adhere to the gaps between the matrix. The formed dense surface can physically encapsulate internal hydrolysable minerals. Meanwhile, the generated C-S-H gel could chemically combine with Ca²⁺. The combined effect of physical encapsulation and chemical combination led to a decrease in electrical conductivity.

The evolution of chromium concentrations in leachates of the pastes with four curing periods is illustrated in Figure 12. The releasing trend of chromium from the composite paste at each curing period was relatively consistent. In the period of the initial leaching, the release rate of chromium in the dry powder increased rapidly and reached to the maximum value of $0.41 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ at the fourth leaching day, which was much faster than the other three cured pastes. After reaching peaks, the chromium leaching rates experienced a rapid decline and reached a stable value at the latter leaching stage. The minimum chromium leaching rate in dry powder, 3-, 7-, and 28-day-cured paste fluctuated around $6.4 \times 10^{-2} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, $4.2 \times 10^{-2} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, $4.0 \times 10^{-2} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, and $3.8 \times 10^{-2} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, separately. The accumulated leaching amount of chromium was 3.6 mg kg⁻¹, 3.0 mg kg⁻¹, 2.6 mg kg⁻¹, and 2.0 mg kg^{-1} in 20 days, corresponding to the curing time of paste for 0, 3, 7, and 28 days, respectively (see Figure 12b). It could be concluded that with the curing time increasing, the leachability of chromium from the composite declined. By analyzing Figure 9, it became known that if the curing time was shorter than 28 days, the dense microscopic morphology cannot be formed. In this case, the leachate cannot be effectively blocked from the Cr-bearing phases. The S/S efficiency of cementitious behavior on Cr leachability was increased continuously with curing time [64]. The generation of this C-S-H gel took place in the pores of the hardened composite pastes' matrix. With the extension of the curing period, the continuously generated C-S-H gel reduced the porosity and hindered the contact of chromium-bearing soluble phases with the solution to elute Cr.



Figure 12. Evolution of (**a**) daily and (**b**) cumulative Cr concentrations in leachates for pastes with different curing times.

3.3.2. Effects of AODS's Dosage on Chromium Leachability

The evolution of chromium leaching ratio of pastes with different AODS dosages is presented in Figure 13. It could be seen that the chromium leaching ratio significantly increased with the increase of AODS dosage. The P5 released more than 10% chromium after 20-day-leaching, while the P3 released only about 5% chromium. When the AODS dosage was lower than 22%, the daily chromium released concentrations from P1 (the entire leaching cycle) and P2 (after the 5th leaching day) were below the detection limit of ICP-OES (0.004 mg/L), so their data were not presented completely in Figure 13b.



Figure 13. Evolution of (**a**) daily and (**b**) cumulative Cr leaching ratio from pastes with different AODS dosages (The daily Cr released concentrations from P1 and P2 were below the detection limit, so their data were not presented completely).

As shown in Figure 10, when the AODS dosage was higher than 44%, the hydration products failed to fill or bond the porosity between the matrix. As cement incorporation increased, large amounts of hydration products were produced after 28-day-curing. Some of the gelling substances, mainly C-S-H gel, filled the pores between the matrix and formed a dense surface.

Sufficient cement incorporation and long enough curing time were two keys to the successful implementation of S/S for chromium in composite pastes. Both of these can increase the hydration rate of the composite material and generate more hydration products. A significant role in the S/S was attributed to C-S-H gel—the primary hydration outcome of the hardened pastes [54]. An increasing amount of C-S-H gel leads to lower permeability. Physical encapsulation of the C-S-H gel prevented the leachate in contact with chromium-bearing phases and decreased the dissolution rate of these phases, and thereby stabilized the chromium.

4. Conclusions

In this study, the cementitious properties of AOD slag and the S/S efficiency of chromium in cement-based composite cementitious materials with various curing conditions were investigated. Some conclusions have been found:

1. The most beneficial AODS mineral phase for cementitious behavior was β -C₂S. The utilization of a higher AODS dosage in composite binders increased the water requirement, prolonged the setting time, weakened the hydration heat, and decreased the strength indexes.

2. Although the AODS possessed limited cementitious properties, it can fulfill the standard of the Grade II SS powder for cement and concrete.

3. With lower AODS dosage and longer curing time, the S/S efficiency for chromium in the composite paste was improved.

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