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Characterization of High-Temperature Hierarchical Porous Mullite Washcoat Synthesized Using Aluminum Dross and Coal Fly Ash

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Abstract: Mixture of aluminum dross (AD) and coal fly ash (CFA) was used to produce high-temperature porous mullite for washcoat application. CFA is the combustion by-product of pulverized coal in a coal-fired power plant, while AD is a waste product produced in secondary aluminum refining. In this study, 80 wt% of AD and 20 wt% of CFA was used to prepare a mullite precursor (MP) via acid leaching and dry-milling. The precursor was coated on a substrate and subsequently fired at 1500 °C. The results showed that the precursor transformed to a hierarchical porous microstructure assembled by large interlocked acicular mullite crystals. The pore structures consisted of large interconnected open pores and small pores. The specific surface area of the mullite washcoat was 4.85 m²g⁻¹ after heating at 1500 °C for 4 h. The specific surface area was compatible with the specific surface area of other high-temperature washcoats.

Keywords: mullite; washcoat; hierarchical; porous; wastes

1. Introduction

High-temperature catalytic processes that aim at creating a more efficient and less polluting chemical process are being used in many applications. For example, emission control of CO, NOx, SOx, hydrocarbons, and carbon particles in internal combustion engines, as well as high-temperature fuel cell and catalytic combustion in power stations which utilizes the catalytic process to promote oxidation reactions of fuel and diminish the formation of unwanted products. The catalyst supports used in these processes basically consist of two components. The first component is a honeycomb monolith ceramic that is made of materials with a low coefficient of thermal expansion (CTE) such as cordierite, aluminum titanate, alumina, silicon carbide, and aluminosilicate mixtures. The second component, a small weight fraction of the catalyst support, is a surface covering of washcoat impregnated with the active catalyst (platinum, palladium, or other noble metals).

The washcoat is the crucial component of a catalyst support. It is where the catalytic reactions take place, as the main function of the washcoat is to provide high contact surface area between the catalyst and reacting gases for high reaction rates. Additionally, the washcoat has to be thermally stable at



working temperatures. A variety of materials that function as a high surface area washcoat such as gamma alumina (γ -Al₂O₃), silica (SiO₂) and titania (TiO₂) have been investigated [1–3]. These high surface area materials are intermediate phases that would undergo further phase transformations upon heating at higher temperatures. The phase transformations would normally transform the materials to high temperature stable phases with a lower specific surface area. For example, Gu et al. [4] reported that mesoporous structure of amorphous silicon dioxide with a high surface area of 1200 m²g⁻¹ could be maintained up to a temperature of 650 °C, but at the annealing temperature of 800 °C, the mesoporous structure was found to collapse completely. Mahlambi et al. [5] reported that amorphous titania exhibited a large surface area (281.5 m²g⁻¹) which decreased to 7.8 m²g⁻¹ when the calcination temperature was increased to 600 °C. Similarly, Suoto et al. [6] reported that γ -Al₂O₃ must be stabilized with alkaline earths or rare earths to slow down the rate at which it was transformed to α -Al₂O₃, because this transformation, which occurs at temperatures around 1000 °C, resulted in a substantial loss of surface area of the washcoat. Generally, these washcoat materials cannot maintain their high surface area microstructures under severe conditions at temperatures above 1000 °C.

One material which appears to be a promising candidate for this high-temperature (>1000 °C) washcoat application is mullite, commonly denoted as $3Al_2O_3 \cdot 2SiO_2$. The advantage of mullite is its ability to produce high thermal stability porous microstructures while exhibiting useful properties such as thermal shock resistance, chemical attack resistance, creep resistance, high heat strength and a low thermal expansion coefficient [7]. Pyzik et al. [8] successfully produced acicular mullite washcoat by a catalytic gas–solid synthesis reaction. The process was based on the formation of fluorotopaz in the presence of silicon tetrafluoride (SiF₄) gas, and subsequently decomposed to form mullite plus free SiF₄ at temperatures above 1000 °C. The catalyzed acicular mullite catalytic filter showed excellent NO_x reduction. However, the downside of this synthesis process is that it needs to be performed in an enclosed and controlled environment because of the SiF₄ gas used, which is very dangerous for human health and the environment.

In this context, the aim of this study is to investigate the feasibility of using aluminum dross (AD) and coal fly ash (CFA) in the production of mullite washcoat via a more environmentally friendly conventional slurry impregnation method. CFA is the combustion by-product of pulverized coal in coal-fired power plant which has a high content of useful silica and alumina, while AD is a high alumina source waste product produced in secondary aluminum refining. The two industrial wastes are suitable as the raw materials for mullite ceramics production. In our previous work, AD and CFA were successfully used to prepare highly crystalline single-phase mullite ceramics [9] and high temperature thermal barrier porous mullite ceramics [10].

2. Materials and Methods

The raw material AD was obtained from a local secondary aluminum refining plant in Rawang, Malaysia, and CFA was obtained from Sultan Salahuddin Abdul Aziz coal-fired power plant in Kapar, Malaysia. Mullite precursor powder was produced according to the procedure reported in our previous study [9]. In the process, 80 wt% of AD was dry-milled with 20 wt% of CFA using a planetary ball-mill (Fritsch Pulverisette 6) for 1 hour at 300 rpm. The mixture was then treated with 2 M hydrochloric acid (HCl) for 1 hour under constant stirring and subsequently filtered off, washed with deionized water, and dried at 100 °C for 2 h.

The obtained powder mixture was made into slurry using deionized water with 35 wt% solid content and dip coated on flat alumina substrates. The loaded substrates were withdrawn and the excess mullite precursor slurry was removed by blowing air. The loaded substrates were then dried at 100 °C for 1 hour and fired at 1200 °C and 1500 °C for 4 h at a rate of 10 °C/min in an electrical box furnace. For identification purpose, the unfired loaded-substrate was named as AS, the fired loaded-substrates were named as MA and MB, corresponding to substrates that were fired at 1200 °C and 1500 °C, respectively. The flow chart of preparing mullite washcoat on alumina substrates is shown in Figure 1.



- U = Dry-milling 80 wt% of AD with 20 wt% of CFA for 1 hour at 300 rpm, leaching with 2 M HCl for 1 hour. Filtering, washing with deionized water, and drying at 100 °C for 2 hours.
- V = Mixing with deionized water at 35 wt% solid content.
- X = Dip coating on alumina substrates, drying at 100 °C for 1 hour.
- $\pmb{Y} =$ firing at 1200 °C for 4 hours at a rate of 10 °C/min.
- Z = firing at 1500 °C for 4 hours at a rate of 10 °C/min.

Figure 1. The preparation of mullite washcoat on alumina substrates.

The chemical composition of the starting materials was determined by energy dispersive X-ray fluorescence (EDXRF) using a Shimadzu EDX-7000. Particle size distributions of the powders were measured with the aid of a laser particle size analyzer (Microtrac X100). Zeiss GeminiSEM 500 Field emission scanning electron microscope (FESEM) was used for the study of the morphology of the starting mullite precursor and the sintered mullite washcoat on alumina substrates. The X-ray diffraction (XRD) pattern at room temperature for the samples were recorded by using PANalytical X'Pert PRO using monochromated CuK α radiation ($\lambda = 1.54184$ Å), and the phase composition was analyzed by Rietveld's refinement method. N₂ physical adsorption–desorption isotherm of the washcoat was obtained with a surface area analyzer (Quantachrome Autosorb-1-C) at 77 K. For the Brunauer–Emmer-Teller (BET) surface area measurement, the washcoat was scraped from the substrate, loaded into a sample cell and degassed at 300 °C for 3 h prior to the analysis. For the calculation of the BET surface area, relative pressure (P/P₀) range of 0.1 to 0.3 was used. The total pore volume was determined from the amount of nitrogen adsorbed at a P/P₀ of 0.995. The pore size distribution was determined by the Barrett–Joyner–Halenda (BJH) method.

3. Results and Discussion

3.1. Characterization of Starting Materials

Figure 2 shows the XRD patterns and the identified crystalline phases of the AD and CFA, which were reported in our previous work [9]. The crystalline phases identified in AD were aluminum (ICDD: 98-006-2866), corundum (ICDD: 98-006-3143), iron oxide (ICDD: 98-008-8471), calcium silicate (ICDD: 98-009-9687), hercynite (ICDD: 98-006-3172) and aluminum phosphide (ICDD: 98-007-9264). The major crystalline phases of CFA were quartz (ICDD: 98-004-0867) and mullite (ICDD: 98-008-0384). There is a characteristic broad diffraction peak between 17° and 30°, centered at 2-theta angle of 22.5° in the XRD pattern, which attributed to amorphous phase in CFA.



Figure 2. XRD patterns showing the crystalline phase identification of two starting materials: (top) aluminum dross, AD and (bottom) coal fly ash, CFA.

Table 1 provides the chemical composition of AD and CFA determined by EDXRF, which from our previous work [9]. The main chemical components observed in AD were Al₂O₃, while the minor components were SiO₂, K₂O, Fe₂O₃, CaO, TiO₂, P₂O₅ and CuO. The chemical components of the CFA were mainly SiO₂ and Al₂O₃ with a smaller amount of other oxides such as K₂O, Fe₂O₃, TiO₂, P₂O₅, CaO and CuO. Table 1 also shows the chemical composition of the mullite precursor powder, which was produced using AD and CFA via the preparation process shown in Figure 1. The Al₂O₃/SiO₂ weight ratio of the mullite precursor powder is 2.48. The value is close to the Al₂O₃/SiO₂ weight ratio of a stable stoichiometric 3:2 mullite (3Al₂O₃·2SiO₂, 71.8 wt% Al₂O₃) which is calculated as 2.55. The right proportion of Al₂O₃ and SiO₂ in the precursor is essential to produce a mullite ceramic with good mineralogy and crystallinity. There were small amounts of impurities such as K₂O, Fe₂O₃, TiO₂, P₂O₅, CaO and CuO still present in the mullite precursor powder. The presence of these impurities can decrease the viscosity of the mullite precursor powder during sintering and thus promotes mullite phase formation [9].

Commenced	Content (wt%)							
Compound	Al ₂ O ₃	SiO ₂	K ₂ O	Fe ₂ O ₃	TiO ₂	P_2O_5	CaO	CuO
aluminum dross (AD)	89.69	5.38	2.37	1.04	0.16	0.28	0.48	0.20
coal fly ash (CFA)	34.48	57.13	3.67	3.46	0.48	0.33	0.32	< 0.01
Mullite precursor (MP)	68.54	27.60	1.56	0.84	0.27	0.21	0.15	0.36

Table 1. Chemical compositions of aluminum dross (AD) and coal fly ash (CFA).

Particle size distributions of the starting materials (CFA, AD) and the mullite precursor are shown in Figure 3. The CFA has a very broad size distribution (2–200 μ m) and a characteristic diameter d50 (average particle size) of 21.83 μ m. The particle size distribution of the AD indicated a narrower size distribution (0.8–88 μ m) and a smaller d50 of 11.58 μ m. The results showed that the combined actions of leaching and dry-milling processes effectively produced mullite precursor with a relatively narrower particle size distribution. A smaller d50 of 10.29 μ m was also recorded for the mullite precursor. Small and homogeneous particles are generally more preferable in washcoat preparation due to their good adherent bond with monolith [11].



Figure 3. Particle size distributions (logarithmic scale on x-axis) of the CFA, AD and mullite precursor powder.

3.2. Sintering and Phase Evolution Characterization

Figure 4 shows the XRD patterns and identified crystalline phases of the mullite precursor powder fired at 1200 °C and 1500 °C. The single crystalline phase identified in the 1500 °C fired mullite precursor powder was mullite. The crystalline phases of 1200 °C fired mullite precursor powder were mullite, corundum, quartz, cristobalite and hercynite. Crystalline phases of the AD such as aluminum, iron oxide, calcium silicate and aluminum phosphide were not detected in the fired mullite precursor powders. This is most likely due to the leaching by HCl treatment, heat-induced decomposition, and phase transformation processes.



Figure 4. XRD patterns show the crystalline phase identification of two mullite precursor powders fired at (top) 1500 °C and (bottom) 1200 °C.

The firing temperature plays a significant role in the mullite mineralogy of fired mullite precursor powder. The mineralogical compositions of the mullite precursor powder fired at 1200 °C and 1500 °C from our previous work [9] are given in Table 2. The result shows that 1500 °C fired mullite precursor powder is solely comprised of mullite crystalline phase. While the 1200 °C fired mullite precursor

powder comprised of several crystalline phases, with mullite (36.7 wt%) as the major phase. It still has a large amount of unutilized crystalline SiO₂ (17.4 wt% quartz, 14.6 wt% cristobalite) and crystalline Al₂O₃ (24.7 wt% corundum). The result concludes that a firing temperature of 1200 °C does not fully combine the crystalline SiO₂ and Al₂O₃ to form mullite.

No.	Firing Temperature	Composition of Mineralogical Phases (wt.%)						
		Mullite	Quartz	Cristobalite	Corundum	Hercynite		
1	1200 °C	36.7	17.4	14.6	24.7	6.7		
2	1500 °C	100	0	0	0	0		

Table 2. Mineralogical compositions of the mullite precursor powder fired at 1200 °C and 1500 °C.

Hercynite, which was originated from AD (Figure 2), was detected in the 1200 °C fired mullite precursor powder but not in the 1500 °C fired mullite precursor powder. Hercynite, in its pure form, is a good refractory material with a high melting temperature (1780 °C). The low melting point of hercynite in Table 2 could be attributed to the small amounts of impurities such as K_2O , P_2O_5 and CaO (Table 1). These impurities have been known for lowering the melting temperature of the ceramic by promoting liquid phase generation during sintering. The low temperature dissolution of hercynite at 1300 °C was also reported by Liu et al. [12].

Figure 5 consists of FESEM micrographs of the mullite precursor coated on substrate (AS) and the resultant products of firing at 1200 °C (MA) and 1500 °C (MB). Figure 5a,b show that the initial mullite precursor was consisted of agglomerates with various particle sizes. After firing at 1200 °C, the mullite precursor transformed to a heterogeneous structure (Figure 5c) with a fraction of the agglomerated particles melted and fused together. Examination of the melted fraction at a higher magnification showed that the microstructure consisted of a glass matrix with small acicular mullite crystals embedded (Figure 5d). On the other hand, sample MB (Figure 5e), which was fired at a higher temperature of 1500 °C, exhibits a three-dimensional hierarchical porous microstructure assembled by large interlocked acicular mullite crystals (Figure 5f). The microstructure of MB has a more evident open porosity compared to MA. The large open pores of MB were well interconnected. Besides the large open pores, small pores were also observed in the microstructure. These small pores were voids between the large interlocked acicular mullite crystals. Generally, the FESEM micrographs showed that although the mullite precursor coated on substrate consisted of agglomerates with various particles sizes (Figure 5a) and has transformed to a heterogeneous structure after being fired at 1200 °C.

Figure 6 shows the schematic diagrams of the mullite washcoat formation mechanism. The formation started in the agglomerated AD and CFA particles. At a firing temperature of about 1200 °C, the SiO₂ in CFA first started to melt and fused with some nearby particles. This is because the SiO₂ that originated from CFA was excessive and associated with impurities such as K_2O , Fe₂O₃, TiO₂, P₂O₅, CaO and CuO. The presence of these impurities has lowered the melting point of the SiO₂ and also decreased the viscosity of the molten liquid phase [13]. At the same time, Al₂O₃ from the AD has begun to diffuse into the SiO₂-rich molten liquid phase and formed an aluminosilicate molten liquid phase. Meanwhile, the small and relatively stable mullite crystals from CFA still remained inside the aluminosilicate molten liquid phase. As the firing temperature increased above 1200 °C, the small mullite crystals acted as seeds and continued to grow at the expense of the aluminosilicate molten liquid phase has all been used up to form large interlocked acicular mullite crystals. The dimensional shrinkage caused by this removal of the aluminosilicate molten liquid phase subsequently rendered a three-dimensional hierarchical porous microstructure.



Figure 5. FESEM micrographs of the surface of the mullite precursor coated on substrate, (**a**) no firing (AS); (**c**) fired at 1200 °C (MA); (**e**) fired at 1500 °C (MB) and the closer views of (**b**) AS; (**d**) MA; (**f**) MB.



Figure 6. Cross-sectional schematic diagrams showing the mullite washcoat formation mechanism, (a) agglomerated AD and CFA particles as mullite precursor; (b) melting and fusing of particles—small acicular mullite crystals embedded in aluminosilicate glass matrix; (c) dimensional shrinkage and formation of hierarchical porous structure with large interlocked acicular mullite crystals; (d) small pores and large interconnected open pores in the microstructure.

3.3. Specific Surface Area and Pore Size Distribution

Figure 7 showed the nitrogen (N₂) physical adsorption–desorption isotherms of the washcoat. The obtained adsorption isotherm was used to calculate the specific surface area, total pore volume, average pore size and pore size distribution. The total pore volume and average pore size were 8.79×10^{-3} cm³g⁻¹ and 7.25 nm, respectively. The BET specific surface area of the mullite washcoat was $4.85 \text{ m}^2\text{g}^{-1}$. This value is considered very low compared with low-temperature washcoat such as γ -alumina (200 m²g⁻¹) and mesoporous silica (1200 m²g⁻¹). However, γ -alumina and mesoporous silica could not maintain their high surface area at above the temperature of 800 °C [4] and 1125 °C [14], respectively. In an application such as catalytic combustion where the temperature range of operation is between 1000 and 1400 °C, a very high surface area washcoat is probably not needed. It was reported that a surface area around 1-10 m²g⁻¹ would be sufficient since both mass transfer limitations and reaction rates are high in the application [15].



Figure 7. Nitrogen's (N₂) physical adsorption–desorption isotherms of washcoat synthesized using aluminum dross and coal fly ash.

The BJH method was used to analyze the pore size distribution as shown in Figure 8. The result showed that the pores of the washcoat were consisted of mesopores and macropores. According to the International Union of Pure and Applied Chemistry (IUPAC) classification [16], pores with widths exceeding about 50 nm are classified as macropores, pores of widths between 2 nm and 50 nm are classified as mesopores, and pores with widths not exceeding about 2 nm are classified as micropores. The adsorption–desorption isotherms (Figure 7) exhibited a hysteresis behavior in the region of relative pressure (P/P₀) between 0.15 and 0.85, this also indicated that the washcoat contained mesopores. A hysteresis appearing in the multilayer range of physisorption isotherms is generally associated with capillary condensation in mesopore structures [16].

A comparison with washcoats from previous studies is shown in Table 3. The comparison showed that the mullite washcoat has a higher specific surface area than the corundum or α -alumina washcoat [6,14] at high temperatures. In addition, the mullite also has a low thermal expansion coefficient compared to α -alumina from our previous study [9]. This showed that the mullite washcoat was more suitable as a high-temperature washcoat compared to α -alumina.



Figure 8. Barrett–Joyner–Halenda (BJH) cumulative pore size distribution (logarithmic scale on the x-axis) of washcoat synthesized using aluminum dross and coal fly ash.

Reference	Raw Materials	Ceramics Product	Synthesis Process	BET Specific Surface Area (m ² g ⁻¹)/Temperature
This work	aluminum dross + coal fly ash	mullite	heating at 1500 °C	4.9/1500 °C
[6]	kaolin	corundum	heating at 1300–1500 °C in a controlled reducing atmosphere	10.5/1300 ° C8.5/1450 ° C3/1500 °C
[14]	boehmite	γ-alumina δ-alumina θ-alumina α-alumina	heating at 450 ° Cheating at 850 ° Cheating at 1000 ° Cheating at 1125 °C	200/450 ° C120/850 ° C50/1000 ° C1/1125 °C
[17]	Aluminum nitrate nonahydrate + Magnesium nitrate hexahydrate	Magnesium aluminate	Precipitation and heating at 1400 °C	7.5/1400 °C
[18]	Aluminum isopropoxide + lanthanum nitrate	lanthanum hexaaluminate	Precipitation and heating at 1400 °C	17/1400 °C
[19]	BaCl ₂ + boehmite	Barium hexaaluminate	Precipitation and heating at 1300 °C	6.6/1300 °C
[20]	Yttrium nitrate + Aluminum nitrate	Yttrium aluminum oxide	Precipitation and heating at 1400 °C	2/1400 °C

Table 3. Comparison of the Brunauer–Emmer-Teller (BET) specific surface area between the present mullite washcoat and other existing washcoats produced from different raw materials.

The specific surface area of the mullite washcoat was also compatible with the specific surface area of other well-studied high-temperature washcoats such as magnesium aluminate, hexaaluminate and yttrium aluminum oxide. Although the specific surface area of the mullite washcoat was slightly lower compared to magnesium aluminate and hexaaluminate, it was within the range of $1-10 \text{ m}^2\text{g}^{-1}$, the minimum requirement suggested by Johansson et al. [15] for catalytic combustion washcoat. It is

important to note that all these commercial high temperature washcoats [17–20] were synthesized from high purity laboratory chemicals and required precise precipitation and heating processes.

4. Conclusions

High-temperature porous mullite washcoat was produced from AD and CFA industrial wastes. The experimental results revealed that at the firing temperature of 1500 °C, mullite precursor (80 wt% AD + 20 wt% CFA) was fully transformed to a solely mullite crystalline phase. The washcoat consisted of a three-dimensional hierarchical porous microstructure assembled by large interlocked acicular mullite crystals. The pore structures were made up of large interconnected open pores and small pores in the forms of mesopores and macropores. The value of the N₂ physisorption BET surface area for the mullite washcoat was 4.85 m²g⁻¹. This relatively high surface area after heating at 1500 °C for 4 h was compatible with the specific surface area of other high-temperature washcoats such as α -alumina, magnesium aluminate, hexaaluminate and yttrium aluminum oxide. Therefore, the mullite washcoat has the potential to be used as washcoat for high temperature applications. In addition, the CFA and AD are environmentally friendly raw materials, and can be used as an alternative to the expensive industrial chemicals.

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