

## Synthesis of phosphate-based binders from aluminum slag for heat-resistant inorganic coatings

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### ABSTRACT

*This study presents the synthesis and optimization of a polyaluminum phosphate (PAP) binder using aluminum slag as the primary source of  $Al^{3+}$ . The binder was synthesized via acid–base reactions between aluminum slag and phosphoric acid ( $H_3PO_4$ ), with varying Al/P molar ratios, temperatures, and stirring speeds. Optimal conditions were identified at 30 wt% slag and 70 wt%  $H_3PO_4$ , 60 °C, and 800 rpm, producing a homogeneous gel with a viscosity of 101 mPa·s, specific gravity of 1.46 g/cm<sup>3</sup>, and excellent adhesion. FTIR analysis confirmed the formation of Al–O–P and pyrophosphate bonds, while thermal and mechanical stability were evaluated under controlled conditions. The results indicate that aluminum slag can be effectively valorized as a raw material for high-performance inorganic binders.*

**Keywords:** Aluminium slag; Poly aluminium phosphate; Inorganic binder; Waste valorization; FTIR analysis; Viscosity.

### 1. INTRODUCTION

- Traditionally, aluminum sources used in the synthesis of phosphate binders include pure aluminum hydroxide or alumina powders. However, these materials are often costly and energy-intensive to produce. In recent years, research has shifted toward sustainable approaches using industrial by-products such as aluminum slag or secondary aluminum dross [7, 8]. Aluminum slag, a by-product generated during secondary aluminum smelting, contains a significant amount of  $Al_2O_3$  and other oxides such as  $SiO_2$ ,  $MgO$ , and  $Fe_2O_3$ . Its valorization not only reduces environmental burden but also provides a low-cost alternative feedstock for binder synthesis [9, 17].

- The utilization of aluminum slag in binder systems not only supports circular economy strategies but also aligns with the growing need to develop cost-effective, eco-friendly materials in the construction and protective coatings industries [10–12]. However, the reactivity and performance of phosphate binders derived from slag are highly dependent on synthesis parameters such as the Al/P molar ratio, temperature, pH, and mixing conditions [13]. Optimization of these factors is essential to ensure the formation of a stable, adhesive, and thermally durable binder.

- This study focuses on the synthesis and optimization of a phosphate-based binder derived from aluminum slag and phosphoric acid, aiming to establish a practical route for transforming industrial waste into valuable functional materials. The influence of key parameters, such as temperature,  $Al^{3+}/PO_4^{3-}$  ratio, and stirring rate, on the viscosity, adhesion, and thermal properties of the binder is systematically investigated using characterization techniques, including FTIR, Viscosity, Density, Adhesion, and SEM-EDS. The outcomes provide insights into the reaction mechanisms and structure-property relationships of PAP binders derived from aluminum slag.

### 2. PROBLEM

#### 2.1. Theoretical foundations

##### 2.1.1. Scientific hypothesis

Inorganic phosphate binders can be synthesized through acid–base reactions between  $Al^{3+}$  sources (e.g., aluminum slag) and phosphoric acid ( $H_3PO_4$ ), forming a polyphosphate network rich

in Al–O–P linkages. Under optimized conditions (temperature, Al/P molar ratio, stirring speed), the reaction produces a homogeneous gel with high thermal resistance and strong adhesion—suitable for heat-resistant coating applications.

### 2.1.2. Chemical reactions and theoretical formulas

The reaction forming the polyphosphate network is as follows:



Reaction forming Al–O–P linkage and polyphosphate chain:



Characteristic bonds identified by FTIR include: asymmetric stretching of  $\text{PO}_4^{3-}$  groups ( $\sim 1051 \text{ cm}^{-1}$ ), pyrophosphate vibrations ( $\sim 909\text{--}930 \text{ cm}^{-1}$ ), and Al–O–P linkages ( $\sim 700\text{--}750 \text{ cm}^{-1}$ ).

## 2.2. Experimental preparation

### 2.2.1. Experimental equipment

The following instruments were used: magnetic stirrer with heating control, analytical balance ( $\pm 0.0001 \text{ g}$ ), Brookfield viscometer, FTIR spectrometer, SEM with EDS, pycnometer, and cross-cut adhesion tester (ASTM D3359).

### 2.2.2. Experimental materials

- Aluminum slag (AS): collected from a secondary smelting plant in Vietnam, dried at  $105 \text{ }^\circ\text{C}$  for 24 h, then ground and sieved ( $<75 \mu\text{m}$ ).

- Phosphoric acid ( $\text{H}_3\text{PO}_4$ , 85%): used as the phosphate source.

- Distilled water: for dilution and reaction processes.

## 3. RESULTS AND DISCUSSION

### 3.1. Effects on the production process

#### 3.1.1. Effect of temperature

The experiment investigating the influence of temperature was conducted using a mixture containing 30% aluminum slag and 85% phosphoric acid, with a stirring speed of 800 rpm. The temperature range studied was from  $60$  to  $90 \text{ }^\circ\text{C}$ . Results indicated that the range of  $60\text{--}70 \text{ }^\circ\text{C}$  was optimal, enabling a stable gelation process. The resulting binder exhibited a viscosity of approximately  $101 \text{ mPa}\cdot\text{s}$  and achieved 100% adhesion, making it suitable for use as a heat-resistant binder.

At higher temperatures ( $80\text{--}90 \text{ }^\circ\text{C}$ ), although the reaction proceeded more rapidly, localized solidification at the bottom of the beaker was observed. This led to measurement errors and compromised product uniformity. The viscosity of the gel in these samples fluctuated irregularly, making subsequent mixing and application difficult to control.

Therefore, the temperature range of  $60\text{--}70 \text{ }^\circ\text{C}$  was selected as the optimal condition for synthesizing the PAP binder.

*Table 1. Effect of Temperature.*

Temperature ( $^\circ\text{C}$ )	Viscosity ( $\text{mPa}\cdot\text{s}$ )	Adhesion, %	Density ( $\text{g}/\text{cm}^3$ )
50	89	85	1.36
60	101	100	1.46
70	120	100	1.47
80	200	92	1.5
90	246	85	1.54

3.1.2. Effect of  $Al^{3+}$  and  $PO_4^{3-}$  concentration

The effect of aluminum slag content (as the  $Al^{3+}$  source) was examined from 5–35 wt% at 60 °C and 800 rpm. At 30 wt%, gelation was optimal, yielding a homogeneous gel with ~101 mPa·s viscosity and 100% adhesion. Higher slag content (35 wt%) caused excessive thickening, while lower content (<10 wt%) led to incomplete reactions and poor adhesion. Thus, 30 wt% slag was identified as the optimal  $Al^{3+}$  source for PAP binder synthesis.

Table 2. Effect of  $Al^{3+}$  Concentration.

$Al^{3+}$ content (%)	Viscosity (mPa.s)	Adhesion, %	Density (g/cm <sup>3</sup> )
5	45	60	1.22
10	63	72	1.25
15	74	81	1.27
20	89	90	1.3
30	101	100	1.46
35	128	100	1.52

The effect of  $PO_4^{3-}$  content (65–90 wt%) was evaluated at 60 °C and 800 rpm. Optimal gelation occurred at 70 wt%, yielding a gel with ~101 mPa·s viscosity, 100% adhesion, and stable structure. Higher acid content (>70%) caused overreaction and instability, while lower content (<65%) led to incomplete polymerization and weak gel quality. Thus, 70 wt%  $PO_4^{3-}$  was optimal for PAP binder synthesis.

Table 3. Effect of  $PO_4^{3-}$  Concentration.

$PO_4^{3-}$ Content (%)	Viscosity (mPa.s)	Adhesion, %	Density (g/cm <sup>3</sup> )
60	72	50	1.22
65	88	65	1.27
70	101	100	1.46
75	96	95	1.35
80	84	80	1.31
85	68	70	1.29

Based on the above findings, a molar Al/P ratio of approximately 0.3 was identified as optimal for the formation of polyphosphate binders from aluminum slag. This ratio provides a balanced condition in which sufficient  $Al^{3+}$  ions are available to coordinate with phosphate groups to form stable polymer networks, without excess acid disrupting the gel structure. When this ratio deviates too far either too low or too high the polymerization process is hindered, leading to precipitation or unstable, low-viscosity gels with poor adhesion and performance.

3.1.3. Effect of stirring speed

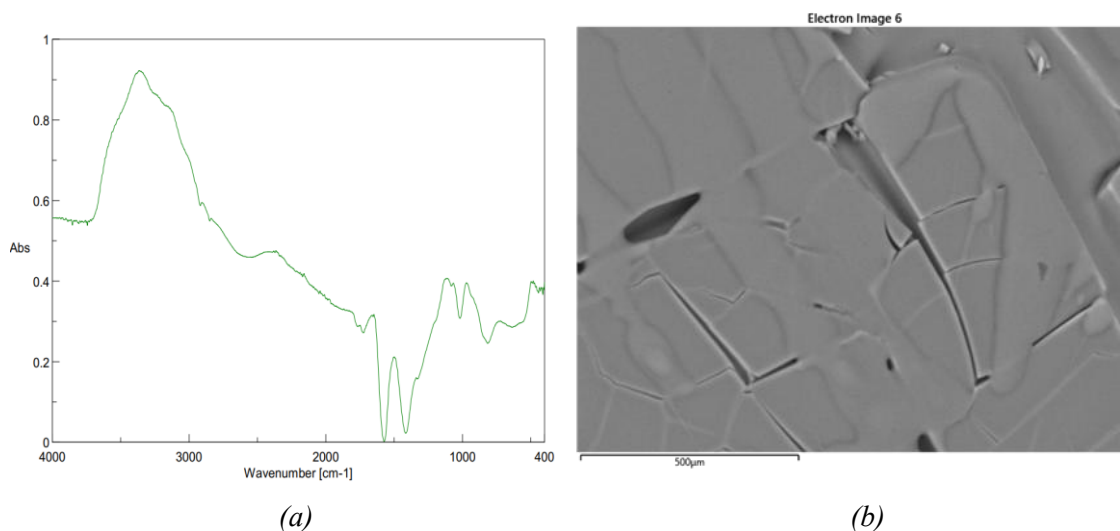
The effect of stirring speed (500–1100 rpm) was studied at 60 °C using 30 wt% aluminum slag. Optimal performance was achieved at 700–800 rpm, producing a uniform gel with ~101 mPa·s viscosity and 100% adhesion. Lower speeds (e.g., 500 rpm) led to incomplete condensation, while excessive speeds (>900 rpm) disrupted gelation. Thus, 700–800 rpm was selected as the optimal stirring range.

Table 4. Effect of Stirring Speed.

Stirring speed (rpm)	Viscosity (mPa.s)	Adhesion, %	Density (g/cm <sup>3</sup> )
500	72	87	1.2
700	91	100	1.34
800	101	100	1.46
900	87	92	1.40
1100	76	81	1.35

### 3.2. Methods, simulation tools

#### 3.2.1. FT-IR analysis



**Figure 1.** FT-IR spectrum (a) and SEM (b) of the synthesized phosphate-binder derived.

The FTIR spectrum of the synthesized phosphate-based binder derived from aluminum slag is shown in figure 1a. Key absorption bands were observed as follows:

A broad band between  $909\text{--}930\text{ cm}^{-1}$ , which is attributed to the asymmetric stretching vibrations of pyrophosphate (P–O–P) groups, indicating the presence of polymerized phosphate chains [1, 6]. A strong peak at around  $1051\text{ cm}^{-1}$ , assigned to the asymmetric stretching of  $\text{PO}_4^{3-}$  units, further confirming phosphate condensation [5]. Peaks in the range of  $700\text{--}750\text{ cm}^{-1}$  are associated with the stretching vibrations of Al–O–P bonds, supporting the formation of a polyaluminum phosphate network [2, 7]. The small shoulder at  $510\text{ cm}^{-1}$  and the band at  $1079\text{ cm}^{-1}$  can be ascribed to Al–O and P–O vibrations, indicating successful integration of aluminum and phosphate species in the inorganic framework [3]. No significant peaks corresponding to organic impurities were detected, affirming the inorganic nature and purity of the binder.

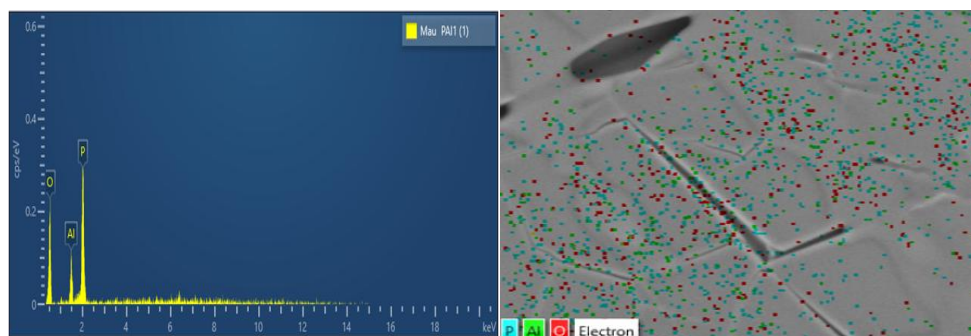
These spectral features are consistent with previously reported spectra for polyaluminum phosphate systems synthesized from either pure alumina or aluminum-rich waste materials [1, 6, 8], confirming the successful formation of a crosslinked phosphate network suitable for heat-resistant applications.

The surface morphology of the polyaluminum phosphate (PAP) binder was analyzed using SEM, as shown in figure 1b. The SEM micrograph reveals that the dried binder forms thin, plate-like structures with uniform distribution across the surface. The observed flake morphology exhibits dimensions in the micrometer range and appears free from visible impurities, indicating a relatively high purity of the synthesized product.

The layered and smooth texture of the gel surface suggests an effective and homogeneous formation of the polyphosphate network, which is consistent with FTIR results indicating the presence of Al–O–P bonds. The absence of cracks and defects further supports the stability and cohesion of the phosphate-based gel, highlighting its potential use as a film-forming agent for heat-resistant coating systems.

These findings confirm that aluminum slag, when properly processed and reacted with phosphoric acid under optimized conditions, can yield a structurally consistent and pure inorganic binder with desirable morphological properties.

3.2.2. EDX elemental composition



Element	Line Type	Weight %	Atomic %
O	K series	55.29	69.87
P	K series	34.92	22.80
Al	K series	9.78	7.33
<b>Total</b>	-	<b>100</b>	<b>100</b>

Figure 2. EDS spectrum of the PAP binder showing Al, P, and O peaks.

The elemental composition of the synthesized phosphate binder was analyzed using Energy-Dispersive X-ray Spectroscopy (EDS), as shown in figure 2. The spectrum reveals distinct peaks corresponding to three main elements: phosphorus (P), oxygen (O), and aluminum (Al)—which are expected in a phosphate-based binder system derived from aluminum slag and phosphoric acid.

Quantitative analysis results are presented in table 5. The predominant elements were oxygen (55.29 wt%) and phosphorus (34.92 wt%), with aluminum accounting for approximately 9.78 wt%. The corresponding atomic percentages were O: 69.87%, P: 22.80%, and Al: 7.33%, confirming a phosphorus-rich and highly oxidized matrix.

These results indicate that the binder consists primarily of phosphate and polyphosphate species, with aluminum successfully incorporated into the phosphate network. The absence of other metallic or non-metallic impurities further supports the high purity of the product and demonstrates the effective removal or suppression of unwanted elements typically present in aluminum slag.

This elemental composition is consistent with the structural inferences drawn from FTIR and SEM analyses, particularly regarding the formation of an Al–O–P bonded framework. The high oxygen and phosphorus content also suggest a highly crosslinked phosphate network, which contributes to the binder's thermal stability and strong adhesion characteristics.

3.3. Preliminary economic assessment and application potential

The utilization of aluminum slag as a raw material for synthesizing phosphate binders not only contributes to the management of industrial solid waste but also significantly reduces material costs compared to conventional aluminum precursors such as aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), aluminum nitrate Al(NO<sub>3</sub>)<sub>3</sub>, or aluminum dihydrogen phosphate Al(H<sub>2</sub>PO<sub>4</sub>)<sub>3</sub>. These commercial chemicals are relatively expensive and require high-purity processing, whereas aluminum slag, a byproduct of aluminum melting processes, is inexpensive, abundant, and environmentally friendly.

Furthermore, the synthesis method used in this study is simple, involving no toxic reagents or specialized equipment, thereby lowering production costs and enhancing the feasibility for practical applications. In contrast to other studies that employ stringent synthesis conditions (e.g., high-temperature calcination, inert atmosphere, or complex purification), the present method operates under ambient conditions and is suitable for laboratory or small-scale industrial deployment.

Moreover, the phosphate-based binder synthesized from aluminum slag demonstrates promising thermal stability and adhesion properties, making it suitable for use in heat-resistant coatings for industrial equipment, furnaces, chimneys, and steel structures. Due to its low-cost raw material and environmental advantage, it also has potential applications in the construction, transportation, and defense sectors where inorganic protective coatings are required. During the experiments, the phosphate binder synthesized from aluminum slag exhibited good stability after curing and no cracking was observed when exposed to ambient humidity. The samples showed no sign of rapid aging, indicating potential for long-term use in harsh environmental conditions.

Regarding thermal stress resistance, coatings using the phosphate-based binder from aluminum slag maintained strong adhesion after heating up to 250–300 °C, indicating acceptable initial thermal tolerance for industrial applications. Although higher temperature testing is pending, the results suggest promising potential for medium-heat equipment.

In terms of mechanical loading, the cured coatings exhibited no visible cracks under mild bending or small impact, demonstrating moderate ductility and the ability to absorb deformation. Additionally, after more than three months of ambient humidity exposure, the coating samples retained structural integrity with no signs of cracking or rapid aging, highlighting their environmental stability and potential for outdoor or humid-environment applications.

#### 4. CONCLUSIONS

In this study, a poly aluminum phosphate (PAP) binder was successfully synthesized from aluminum slag and phosphoric acid under optimized conditions: 30% aluminum slag, 70% H<sub>3</sub>PO<sub>4</sub>, at 60–70 °C with a stirring rate of 700–800 rpm. The resulting binder exhibited high viscosity (~101 mPa·s), excellent adhesion (100%), and stable rheological behavior.

FTIR, SEM, and EDX analyses confirmed the formation of a stable Al–O–P polymeric network with a relatively pure microstructure. After curing, the coating demonstrated good thermal stability and remained intact under ambient conditions.

These findings highlight the potential of utilizing industrial waste for developing high-performance inorganic binders suitable for heat-resistant coating applications.

Key parameters such as viscosity, specific gravity, and adhesion strength were measured in triplicate for each test condition. The results showed minimal variation, with standard deviations below 5%, confirming good repeatability and high reliability of the experimental findings. This suggests the synthesis process is stable and reproducible, supporting its potential for practical application.

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## TÓM TẮT

### Tổng hợp chất kết dính trên nền phosphate từ xỉ nhôm cho lớp phủ vô cơ chịu nhiệt

Nghiên cứu này trình bày quy trình tổng hợp và tối ưu hóa chất kết dính poly aluminum phosphate (PAP) từ xỉ nhôm – một phế thải công nghiệp giàu  $Al_2O_3$ . Phản ứng axit-bazo giữa xỉ nhôm và axit photphoric ( $H_3PO_4$ ) được thực hiện ở các điều kiện khác nhau về tỷ lệ mol Al/P, nhiệt độ và tốc độ khuấy để khảo sát ảnh hưởng đến độ nhớt, độ bám dính và cấu trúc gel. Kết quả chỉ ra rằng điều kiện tối ưu là 30% xỉ nhôm, 70%  $H_3PO_4$ , nhiệt độ 60–70 °C và tốc độ khuấy 800 vòng/phút. Sản phẩm thu được là gel đồng nhất, có độ nhớt cao (~101 mPa·s), tỷ trọng 1.46 g/cm<sup>3</sup> và độ bám dính 100%, đáp ứng tốt yêu cầu làm lớp phủ chịu nhiệt. Phân tích FTIR xác nhận sự hình thành liên kết Al–O–P và chuỗi polyphosphate, trong khi ảnh chụp SEM và phổ EDS cho thấy cấu trúc vi mô đồng nhất và độ tinh khiết cao. Kết quả nghiên cứu khẳng định tiềm năng tái sử dụng xỉ nhôm để sản xuất chất kết dính vô cơ hiệu suất cao, góp phần thúc đẩy phát triển vật liệu thân thiện môi trường.

**Từ khoá:** Xi nhôm; Chất kết dính phosphate; Lớp phủ vô cơ; FTIR; Độ nhớt; Tái sử dụng chất thải.