VALORIZATION OF PHOSPHORUS SLAG FROM TANG LOONG FOR SUSTAINABLE CONSTRUCTION AND INDUSTRIAL APPLICATIONS

PhamThi Minh[1], Nguyen Van Chien[2],
Dam Xuan Thang[3,*]

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ABSTRACT

This study investigates the recycling of phosphorus slag from the Tang Loong Industrial Park (Lao Cai, Vietnam) as a secondary mineral resource for building material applications. Large volumes of phosphorus slag generated from yellow phosphorus production pose significant environmental challenges, while the slag itself is rich in calcium-silicate phases (CaO 50 - 52%, SiO_2 37 - 40%) with a specific gravity of approximately 2.75g/cm^3. After purification via a combined washing and gravity separation process, the resulting slag powder exhibited improved quality and was evaluated in multiple product systems. In coating formulations, the slag effectively replaced mineral fillers at a slag/resin ratio of 1.25/1, achieving high opacity and strong adhesion. In HDPE-based composites, the addition of 5 - 20wt% slag improved tensile strength and elongation at break. In wall putty applications, the slag successfully replaced conventional mineral aggregates while achieving adhesion values close to or meeting the requirement of TCVN 7239:2014. These results demonstrate that properly treated phosphorus slag can serve as an effective alternative to natural mineral powders across different material systems, offering a practical route for large-scale valorization of industrial waste while reducing environmental impact and enhancing sustainable resource utilization.

Keywords: *Phosphorus slag; recycling; mineral filler; HDPE composites; coatings; wall putty; sustainable materials.*

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[1] Electric Power University, Vietnam
[2] Institute of Chemistry, Vietnam Academy of Science and Technology, Vietnam
[3] Faculty of Chemical Technology, Hanoi University of Industry, Vietnam
[*] Email: thangdx@haui.edu.vn

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1. INTRODUCTION

The Tang Loong Industrial Park in Lao Cai is currently under considerable environmental pressure due to the large volume of phosphorus slag discharged from yellow phosphorus production [1, 2]. For each ton of elemental phosphorus (P_4) produced, approximately 8 - 10 tons of slag are generated, leading to substantial annual solid waste accumulation in the region. This slag is discharged from electric furnaces at high temperature and rapidly quenched, producing a predominantly glassy calcium–silicate-rich material with potential for use as a supplementary mineral resource in construction-related applications [1, 2].

In Lao Cai, yellow phosphorus is produced mainly by electric furnace technology using apatite ore, silica-bearing materials, and carbonaceous reducing agents, which results in the generation of large quantities of calcium–silicate-rich slag [23]. Phosphorus slag from Lao Cai is generally grayish-white and mainly composed of CaO and SiO_2, together with smaller amounts of P_2O_5, F^-, Al_2O_3, and Fe-containing phases, which strongly influence its subsequent processing and reuse potential [2-5]. Such a composition makes phosphorus slag a potentially valuable secondary mineral resource for building materials, fillers, and cementitious systems [3, 6, 7].

In Vietnam, considerable efforts have been devoted to valorizing this industrial by-product, and previous studies have demonstrated its potential use in ceramic products, cementitious materials, unfired bricks, and other construction-related systems [7-11]. At the international level, phosphorus slag has been extensively investigated as a supplementary cementitious material and as a reactive component in alkali-activated or blended binder systems [1, 3, 5, 6, 12]. However, its broader utilization in cement-based materials remains limited because residual phosphorus- and fluorine-bearing species can retard cement hydration, prolong setting time, and reduce early-age strength development [1, 3, 6, 12]. Recent mechanistic studies have further confirmed that phosphorus slag delays the precipitation of C–S–H and portlandite, thereby suppressing the early hydration of silicate phases in cement [3, 12].

Despite these limitations, phosphorus slag still shows promising compatibility in non-structural and semi-structural applications, particularly after suitable treatment. It has been evaluated as a mineral filler or functional additive in asphalt binders, asphalt mixtures, and related particulate-filled systems [13-18]. Similar valorization trends have also been widely reported for other industrial slags used in asphalt pavements, further supporting the feasibility of slag-based mineral fillers in road materials [19]. In addition, finely processed phosphorus slag has been reported to exhibit improved physicochemical performance, including enhanced reactivity, modified fresh-state behavior, and increased later-age contribution when incorporated into blended cement systems [3, 5, 12]. These features suggest that, beyond conventional cement replacement, phosphorus slag may also be suitable for applications requiring mineral fillers and functional particulate phases, provided that its impurities and particle characteristics are properly controlled.

Another important motivation for phosphorus slag utilization is the increasing pressure on natural mineral resources in Vietnam, particularly mineral fillers and construction aggregates. When processed to appropriate fineness and stability, phosphorus slag has demonstrated favorable performance in cementitious, filler-containing, and blended construction-related systems [5, 7, 20, 21]. Comparable resource-saving and sustainability benefits have also been reported for other metallurgical slags, such as electric arc furnace slag used in geopolymer and construction materials [22]. Its reuse can therefore simultaneously alleviate waste disposal pressure and reduce dependence on conventional raw materials, contributing to both environmental protection and sustainable resource utilization [1, 11].

In addition, the demand for mineral fillers and construction-related materials in Vietnam remains consistently high across sectors such as plastics, coatings, cement, and building materials. This further highlights the strong potential of phosphorus slag as an alternative mineral resource for diverse industrial applications.

Given these considerations, although phosphorus slag from Tang Loong represents a major industrial waste stream, its current utilization in Vietnam is still concentrated mainly in road-related and other relatively low-value applications, while its broader use in higher-value material systems remains limited [7-11, 23]. A key technical barrier is the persistent presence of soluble or reactive species, especially phosphorus- and fluorine-containing compounds, which can interfere with hydration, workability, and long-term material performance [1, 2, 3, 6, 12]. In particular, recent studies have shown that increasing slag fineness may improve activity, but can also intensify specific physicochemical effects, including hydration retardation and flocculation-related changes in fresh-state behavior [3, 5, 12].

In this context, a clean and cost-effective treatment strategy is required to expand the utilization of phosphorus slag in higher-value material systems. In particular, reducing harmful soluble species and tailoring particle characteristics are essential for extending its application in fillers, coatings, putty, and polymer-based composites, as well as selected cementitious materials. Therefore, this study aims to address the key technical and environmental barriers associated with phosphorus slag reuse through a controlled physical treatment route involving washing, grinding, and gravity separation, with subsequent evaluation in several filler-containing product systems [2, 7]. The treated slag was subsequently evaluated in three product categories, namely coatings, HDPE-based composites, and putty formulations. The results are expected to contribute to a rational utilization strategy for phosphorus slag, simultaneously mitigating environmental impacts and promoting sustainable resource development.

2. MATERIALS AND METHODS

2.1. Materials - Chemicals

Phosphorus slag was collected from a yellow phosphorus production facility in Lao Cai Province, Vietnam. The as-received slag was a grayish-white calcium-silicate-rich by-product generated during electric furnace production. It had an average particle size of approximately 2mm, a specific gravity of about 2.75, and a moisture content below 1wt%. The slag was mainly composed of CaO-SiO_2-based phases, with CaO and SiO_2 accounting for more than 90% of the total composition. Minor amounts of carbonaceous particles, metallic inclusions, soluble P_2O_5, residual apatite, and other reactive impurities were also present.

Industrial-grade CaO, $Ca(OH)_2$, $NaOH$, and Na_3PO_4 were supplied by commercial chemical suppliers in

China. Polyacrylamide (PAM), acrylamide-based flocculants, protein-based flocculants, and bentonite were used as commercial water-treatment additives without further purification.

For coating preparation, acrylic polyol resin HSU1908 was supplied by A & P Industrial Resin Co., Taiwan, with a solid content of approximately 55wt% and a hydroxyl content of about 0.9wt% based on resin solids. Desmodur N75 polyisocyanate curing agent was supplied by Bayer, Germany, with a solid content of approximately 75 wt% and an NCO content of 16 - 17wt%. Commercial-grade xylene was supplied from China. R4322 styrene-acrylic emulsion was supplied by Synthomer, with a solid content of 49 ± 1wt% and pH of 8.0 - 9.5. TiO_2 R902 rutile pigment was supplied by DuPont, while AM-95 and the thickener were supplied by Dow Chemical.

For composite and putty preparation, high-density polyethylene (HDPE) imported from South Korea was used, with a density of 0.937g/cm^3 and a melt flow index of 1.20g/10min at 190°C/2.16kg. Thai Binh white cement PCW30 and But Son gray cement PCB30 were used as cementitious binders. Lime powder, metakaolin, talc powder, and DA1120 redispersible polyvinyl alcohol powder from Taiwan were used as commercial-grade additives for wall putty formulations.

2.2. Methods

2.2.1. Slag purification procedure

The phosphorus slag was purified through a physical treatment route including wet screening, gravity separation, drying, and grinding. This process was designed to exploit the density differences among amorphous silicate particles, metallic inclusions, and carbonaceous impurities. The as-received slag was first classified by wet vibrating screening and then processed on a shaking table to enrich the silicate fraction while removing high-density metallic particles and low-density carbonaceous matter.

To improve the removal of soluble and suspended impurities, the process was integrated with a water treatment system. Calcium oxide (CaO) and slaked lime [$Ca(OH)_2$] were used to precipitate phosphorus-containing species, while protein-based flocculants, polyacrylamide (PAM), acrylamide derivatives, and bentonite were applied to enhance sedimentation and solid-liquid separation. The purified slag was subsequently dried and ground to a particle size below 74µm for further use. Similar physical purification strategies have previously been reported as effective for improving phosphorus slag quality [7].

2.2.2. Characterization methods

The chemical composition and microstructure of phosphorus slag were analyzed by X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDX). XRF was used for elemental analysis, XRD for phase identification, SEM for morphology observation, and EDX for elemental mapping. These techniques have been widely applied in previous studies on phosphorus slag and related mineral materials.

After purification, the slag powder and the prepared products were evaluated in terms of particle size distribution, specific gravity, moisture content, compressive strength, flexural strength, and water absorption. The tests were carried out according to relevant Vietnamese standards, including QCVN 16:2017/BXD, TCVN 6016, and TCVN 8264.

2.2.3. Preparation and formulations of product samples

The treated phosphorus slag powder was used as a mineral filler in three representative product systems, including coating formulations, wall putty, and HDPE-based composites. The main formulations used for sample preparation are summarized in Tables 1 ÷ 3 to improve the clarity and reproducibility of the experimental procedure.

For coating applications, the treated slag was incorporated into both solvent-based acrylic polyol/polyisocyanate coatings and water-based alkali-resistant primer systems. In the solvent-based coating, HSU1908 acrylic polyol resin and Desmodur N75 curing agent were used at an OH/NCO molar ratio of 1/1, while the slag/resin mass ratios of 0/1, 0.5/1, 0.75/1, 1.0/1, 1.25/1, and 1.5/1 were investigated. In the water-based primer, R4322 styrene-acrylic emulsion, TiO_2 R902, treated slag powder, AM-95, thickener, and water were used as the main components. The corresponding coating formulations are shown in Table 1.

Table 1. Formulations of phosphorus slag-containing coating systems

Coating system	Component	Amount or ratio
Solvent-based acrylic polyol coating	HSU1908 acrylic polyol resin	1.00 part resin basis
	Desmodur N75	OH/NCO = 1/1
	Xylene	As required

	Treated phosphorus slag powder	0 - 1.5 parts per 1 part resin
Water-based alkali-resistant primer	R4322 styrene-acrylic emulsion	500g
	Distilled water	25g
	TiO_2 R902	25g
	Treated phosphorus slag powder	400g
	AM-95	4g
	Thickener	2g

Wall putty samples were prepared as dry-mix cementitious formulations using treated P_4 slag/talc as the main mineral filler. Ten formulations were designed per 1000g of dry product, including five exterior-grade samples based on black cement and five interior-grade samples based on white cement. The dry components were homogenized before water addition and subsequent testing according to TCVN 7239:2014. The detailed formulations are presented in Table 2. This formulation design is consistent with the original wall putty report, where treated P_4 slag/talc was used as the major filler phase and the samples were evaluated for fluidity, setting behavior, water retention, adhesion strength, and relative surface hardness.

Table 2. Formulations of experimental wall putty samples, calculated per 1000g dry product

Component	**CT1**	**CT2**	**CT3**	**CT4**	**CT5**	**CT6**	**CT7**	**CT8**	**CT9**	**CT10**
Black cement PC30	250	225	200	175	200	–	–	–	–	–
White cement	–	–	–	–	–	360	324	288	252	200
Treated P4 slag + talc	715	743	743	743	710	655	691	691	691	715
Lime	0	25	50	75	75	0	36	72	108	70
MK	28	0	0	0	2	36	0	0	0	2
Na_3PO_4	–	–	–	–	5	–	–	–	–	5
DA1120	7	7	7	7	8	9	9	9	9	8
Total	1000	1000	1000	1000	1000	1000	1000	1000	1000	1000

HDPE/phosphorus slag composites were prepared with slag contents of 0, 5, 10, 15, and 20wt% based on the total composite mass. The formulations are shown in Table 3. The treated slag powder was incorporated into the HDPE matrix by melt blending, followed by hot pressing to obtain composite sheets for mechanical testing.

Table 3. Formulations of HDPE/phosphorus slag composites

Sample	**HDPE (wt%)**	**Treated phosphorus slag powder (wt%)**
HDPE	100	0
HDPE/5% slag	95	5
HDPE/10% slag	90	10
HDPE/15% slag	85	15
HDPE/20% slag	80	20

The resulting products were evaluated for their physical and mechanical performance in accordance with relevant national and international standards, including TCVN 7239:2014 for wall putty, TCVN 8789:2011 and ISO 2409 for coating adhesion and durability, and TCXDVN 272:2002 for HDPE pipe-related materials. The performance of slag-containing products was then compared with that of corresponding materials prepared using conventional mineral fillers.

3. RESULTS AND DISCUSSION

3.1. Quality of processed phosphorus furnace slag product

After washing, gravity separation, and grinding, the phosphorus slag was converted into a grayish-white powder with a relatively uniform particle distribution, as shown in Figure 1a. SEM observation (Figure 1b) revealed that the treated particles mainly exhibited irregular, angular, and plate-like morphologies, which are typical of mechanically fractured calcium-silicate-rich slag particles. The higher-quality SEM image further shows a rough particle surface with fine adhered fragments, indicating that grinding and physical separation produced fractured particles with increased surface irregularity. This morphology is consistent with previous reports on wet-milled and physically treated phosphorus slag, in which particle refinement improves particle-size uniformity while preserving the angular and fractured nature of the slag matrix [5, 12].

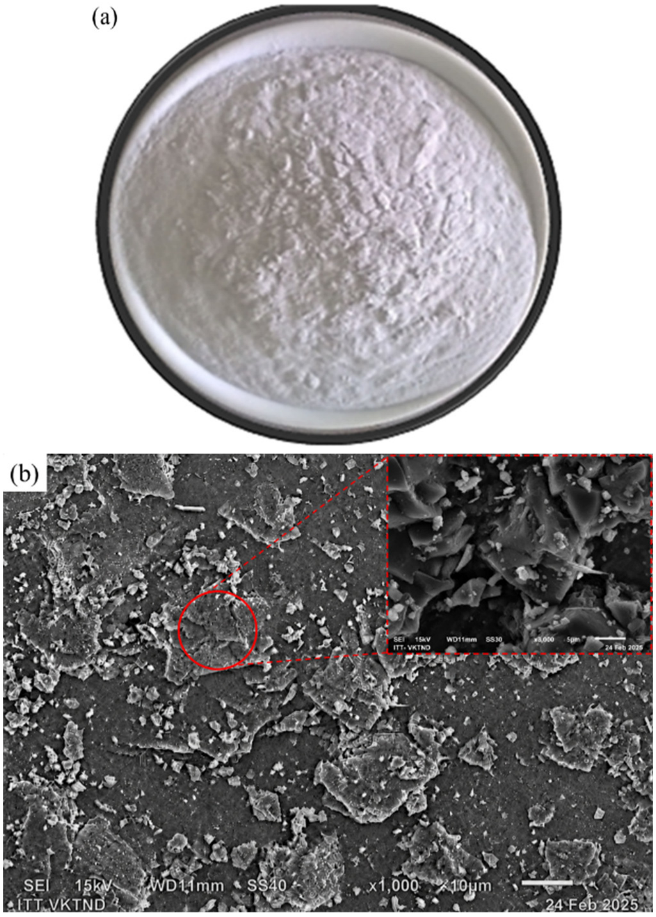

Figure 1. Morphology of slag sample after treatment

(a- Powder sample; b- Powder surface morphology)

The XRD pattern of the treated sample (Figure 2), together with the compositional data in Table 4, indicates that the treated slag was dominated by a calcium-silicate-rich structure with markedly reduced signals associated with impurity-bearing phases compared with the original slag. In particular, peaks attributable to phosphorus- and metallic-bearing components were weakened after treatment, suggesting that the washing-gravity separation process effectively reduced soluble and dense impurity fractions. This interpretation agrees with previous studies showing that phosphorus slag commonly contains residual P- and F-bearing species that can adversely affect material performance and therefore should be reduced prior to further utilization [3, 6, 12].

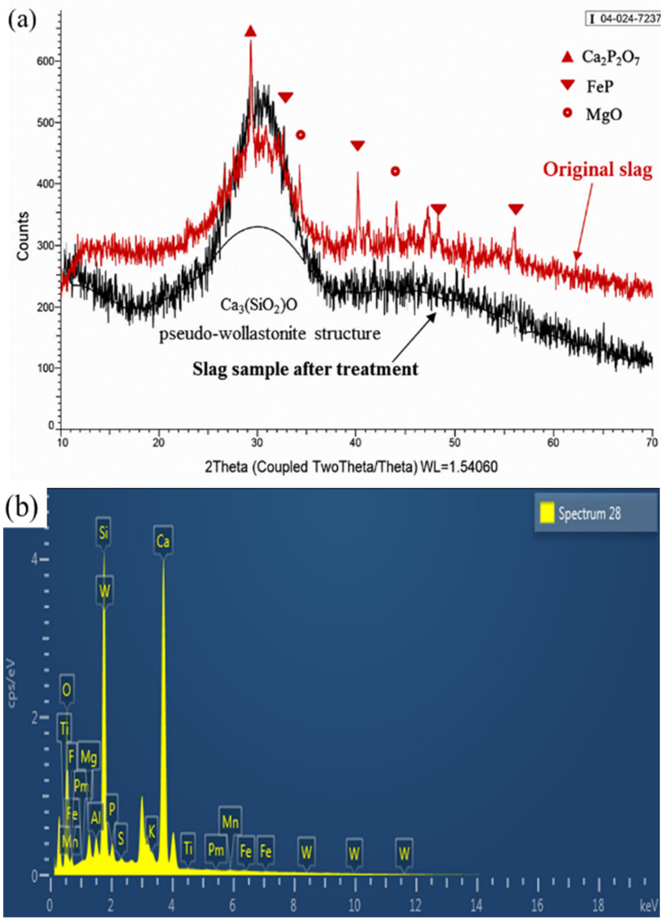

Figure 2. XRD and EDX characterization of phosphorus slag: (a) XRD patterns before and after treatment; (b) representative EDX spectrum of the treated slag

To further support the elemental characterization of the treated phosphorus slag, SEM–EDX analysis was performed on representative areas of the purified powder. A representative EDX spectrum is presented in Figure 2b, while the average elemental composition calculated from Spectrum 28 is summarized in Table 4. The EDX results confirm that the treated slag is mainly composed of O, Ca, and Si, with average contents of approximately 46.84, 29.96, and 13.78wt%, respectively. This result is consistent with the XRD pattern and confirms the calcium-silicate-rich nature of the purified slag. Minor amounts of F and P were also detected, with average contents of approximately 3.70 and 1.09wt%, respectively, indicating the presence of residual fluorine- and phosphorus-bearing species after treatment. However, their relatively low contents, together with the decreased dissolved P_2O_5 content shown in Table 5, suggest that the washing-gravity separation process effectively reduced impurity levels and improved the suitability of the treated slag as a mineral filler.

Table 4. Average EDX elemental composition of the treated phosphorus slag

Element	Average weight (%)	Average atomic (%)	Interpretation
O	46.84	64.67	Main oxide/silicate matrix
Ca	29.96	16.52	Calcium-rich phase
Si	13.78	10.84	Silicate phase
F	3.70	4.31	Residual fluorine-bearing species
Mg	1.46	1.33	Minor oxide component
P	1.09	0.78	Residual phosphorus-bearing species
Al	0.99	0.81	Minor aluminosilicate component
K	0.36	0.21	Minor alkali component
S	0.30	0.21	Trace sulfur-containing species
Fe	0.22	0.09	Trace metallic/oxide impurity
Mn	0.18	0.07	Trace impurity
Ti	0.08	0.04	Trace oxide component

Note: *Semi-quantitative EDX values were obtained from representative Spectrum 28; uncertain trace elements were excluded.*

The physicochemical results in Table 5 further confirm the improvement in product quality after treatment. The whiteness increased from ≥ 80 to ≥ 85, while the dissolved P_2O_5 content decreased from ≤ 1.5% to ≤ 0.10%. At the same time, the total content of metal oxide impurities was reduced from 5.0 - 7.0% to 1.5 - 3.0%, indicating that gravity separation was effective in removing high-density impurity phases. These changes are important because previous studies have shown that impurity removal and particle refinement can significantly improve the physicochemical stability and application potential of phosphorus slag in cementitious and filler-based systems [3, 5, 12].

Table 5. Chemical composition of phosphorus slag samples before and after treatment

No.	Main indicator name	Processing values		Limestone powder (comparative value)
		Before	After	
1	Whiteness	≥ 80	≥ 85	> 95
2	Density, g/cm^3	≈ 2.57	≈ 2.57	≈ 2.7
3	Ca as CaO, %	38 - 45	38 - 45	50 ÷ 55
4	Si as SiO_2, %	38 - 45	38 - 45	< 0.20
5	P_2O_5, % dissolved	≤ 1.5	≤ 0.10	-
6	F, % dissolved	≤ 0.5	≤ 0.5	-
7	Pb, mg/kg (ppm)	≤ 2.0	≤ 2.0	-
8	Cd, mg/kg (ppm)	≤ 0.3	≤ 0.3	-
9	As, mg/kg (ppm)	≤ 0.2	≤ 0.2	-
10	Total Fe_3O_4, MnO, MgO, TiO_2 (%)	5.0 - 7.0	1.5 - 3.0	-
11	Safe natural radioactivity (I)	≈ 0.85	≈ 0.7	-

Although small amounts of phosphorus and fluorine may still remain in the treated material, they are expected to be largely incorporated within or associated with the calcium-silicate-rich matrix, thereby reducing their mobility and potential adverse effects. Similar observations have been reported in studies on phosphorus slag hydration and workability, where the residual effects of P and F were strongly dependent on their amount, fineness, and distribution in the slag structure [5, 6].

Overall, the treated slag powder met the basic technical requirements for replacing conventional mineral fillers in several product systems. Its improved whiteness, reduced impurity level, dominant Ca–Si–O composition confirmed by SEM-EDX, and stable physicochemical characteristics support its potential use in coatings, polymers, putty formulations, and other non-structural material applications [3].

3.2. Material/Phosphorus Slag Preparation

3.2.1. Paint preparation using phosphorus slag powder

Mineral fillers are widely used in coating formulations to improve covering ability, adjust rheological behavior, and reduce formulation cost. In this study, phosphorus slag powder was evaluated as a potential alternative filler for both solvent-based and water-based paint systems. A reference composition range for oil-based and water-based paints is presented in Table 6.

Table 6. General formulation for oil-based and water-based paint systems

No.	Main raw material category	% weight	
		Oil paint	Water paint
1	Resin (film former)	20 - 25	30 - 40
3	Additives	0.1 - 2	12 - 17
3	Pigments	3.0 - 10	40 - 50
4	Fillers	30 - 40	
5	Solvents	20 - 25	20 - 30

For the solvent-based coating system, a polyurethane formulation based on HSU 1908 acrylic polyol resin and Desmodur N75 polyisocyanate curing agent was selected, with an OH/NCO molar ratio of 1:1. The treated phosphorus slag powder, obtained after washing, drying, and grinding, was incorporated as a filler at different slag/resin mass ratios of 0/1, 0.5/1, 0.75/1, 1.0/1, 1.25/1, and 1.5/1. Similar formulation strategies have previously been reported for filler-containing systems using finely processed phosphorus slag or related mineral powders after impurity reduction and particle refinement [1, 3].

The coating was prepared by first dispersing phosphorus slag powder into HSU 1908 resin diluted with xylene under high-speed stirring for 1h. The N75 curing agent was then added and mixed for 5 min. The resulting mixture was applied to test substrates and allowed to air dry, producing coating films with a thickness of approximately 30µm.

The optical opacity and adhesion of the prepared films were evaluated by UV-Vis spectroscopy and cross-hatch adhesion testing (Elcometer 1542), respectively, following ISO 2409. As shown in Table 7, adhesion and opacity improved progressively with increasing slag content up to a slag/resin ratio of 1.25/1. At a higher ratio of 1.5/1, the adhesion score decreased, which may be associated with excessive filler loading and reduced interfacial cohesion within the coating film. The improvement in opacity may be attributed to the presence of finely dispersed calcium-silicate-rich slag particles, which enhance light scattering and filler efficiency. These results indicate that a slag/resin ratio of 1.25/1 provided the best balance among opacity, adhesion, and filler utilization efficiency.

Table 7. Effect of phosphorus slag powder content on paint film adhesion

Slag powder/resin ratio	Adhesion, score	Opacity, %
0.0/1	1	95
0.5/1	1	96
0.75/1	1	97
1.00/1	1	98
1.25/1	1	99
1.50/1	2	99

The coating prepared at the slag/resin ratio of 1.25/1 was further evaluated for protective application on steel substrates. The physicochemical properties listed in Table 8 show that this formulation satisfied the technical requirements of TCVN 8789:2011 for coating systems used in steel structure protection. In addition, a water-based alkali-resistant primer was formulated using R4322 resin and phosphorus slag at the same 1.25/1 ratio. After neutralization and additive incorporation, the product met the technical requirements of TCVN 8652:2012, as summarized in Table 9. Overall, these findings suggest that treated phosphorus slag can function effectively as a partial mineral filler in both solvent-based and water-based coating systems, thereby contributing to reduced formulation cost and broader resource utilization.

Table 8. Some physicochemical properties of HSU1908/N75/slag powder coating for steel structure protection

No.	Properties	Unit	Determined values	Value according to TCVN 8789:2011	
				Priming	Intermediate coat
1	Dry Content	%	83	≥ 50	≥ 60
2	Fineness	µm	≤ 10	≤ 30	≤ 50
3	Adhesion	score	1	1	1
4	Impact Strength	kG.cm	90	≥ 45	≥ 45

Additionally, a water-based alkali-resistant primer was formulated using R4322 resin and phosphorus slag at a 1.25/1 ratio. The product, after neutralization and additive incorporation, met technical standards TCVN 8652:2012 (Table 9).

Table 9. Technical Specifications of the Alkali-Resistant Primer

No.	Indicator name	Unit	Test standard	Measured value	Required value
1	Drying Time				
	- Surface dry	hour	TCVN 2096-1993	0.5	≤ 1[a]
	- Full dry	hour		2	≤ 3[a]
2	Fineness	µm	TCVN 2091:2008	25	≤ 30[a]
3	Adhesion	score	ISO 2409	1	≤ 1[a]
4	Alkali Resistance	hour	TCVN 8653-3:2024	≥ 240	≥ 240[a]
5	Water Resistance	hour	TCVN 8653-2:2024	≥ 480	≥ 480[a]
6	Water Absorption	ml/m^2	TCVN 8652:2020	4	≤ 8[a]
7	Relative Hardness	-	TCVN 8789:2011	0.24	≥ 0.20

Note: "a": Required level according to TCVN 8652:2020. All coating films were naturally dried for 7 days before testing.

3.2.2. Preparation of HDPE/phosphorus slag composite material

In addition to coating formulations, phosphorus slag powder was also investigated as a filler for thermoplastic composite systems. Preliminary trials were first carried out on PVC sheets and PE films, indicating that finely processed phosphorus slag powder could be incorporated into polymer matrices without causing obvious processing instability (Figure 3). Based on these initial observations, the study was extended to HDPE-based composite materials intended for corrugated electrical conduit applications (Figure 4), in line with the direction previously reported for phosphorus slag/polymer composite utilization [24].

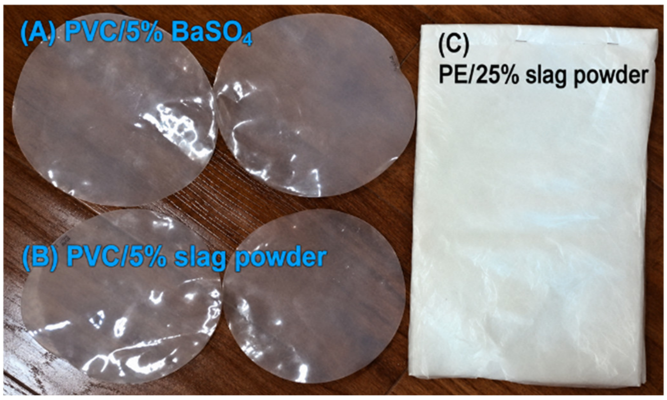

Figure 3. PE films and PVC/slag powder sample

Figure 4. HDPE/slag powder composite corrugated pipe

The HDPE/phosphorus slag composite was prepared by melt blending in a Haake internal mixer (Germany). The mixing process was conducted at 180°C for 5 min with a rotor speed of 50rpm. After melting and homogenization, the blend was removed and hot-pressed using a Toyoseiki hydraulic press (Japan) at 185°C for 2 min under a pressure of 5MPa to form flat sheets with a thickness of 1.0 - 1.2mm. The sheets were then cooled in ambient air and conditioned at room temperature for 24h prior to mechanical testing.

The experimental formulations demonstrated that the addition of fine phosphorus slag powder (≤ 15μm) significantly influenced the mechanical performance of the HDPE composites. As shown in Table 10, when the slag content increased from 0 to 5wt%, both tensile strength and elongation at break improved markedly compared with neat HDPE. At this loading level, the tensile strength increased from 28.09 to 33.86MPa, while the elongation at break increased from 328.51% to 469.64%. These results suggest improved stress transfer and interfacial interaction between the polymer matrix and the dispersed slag particles. Similar trends have been reported in previous studies on polymer systems filled with yellow phosphorus slag, in which moderate filler loading improved the balance between strength and ductility [24].

Table 10. Mechanical properties of HDPE/phosphorus slag composite material

No.	Sample	Elastic modulus, MPa	Tensile strength, MPa	Elongation at break, %
1	HDPE	513.34 ± 5.35	28.09 ± 0.13	328.51 ± 4.72
2	HDPE 5 % slag	459.55 ± 6.18	33.86 ± 0.14	469.64 ± 6.98
3	HDPE 10 % slag	370.48 ± 4.49	28.73 ± 0.18	428.45 ± 5.12
4	HDPE 15 % slag	337.25 ± 3.91	27.14 ± 0.20	420.12 ± 4.51
5	HDPE 20 % slag	312.29 ± 5.57	26.93 ± 0.24	339.26 ± 4.64

At intermediate filler contents of 10 - 15wt%, the elongation at break remained higher than that of unfilled HDPE, indicating that the slag particles were still relatively well dispersed and capable of contributing to ductility. However, the elastic modulus decreased progressively with increasing filler content, suggesting that the treated slag acted not only as a rigid particulate phase but also affected the continuity and deformability of the polymer matrix. This tendency is consistent with previous observations in Ca-Si-based and industrial mineral-filled polymer composites [24].

When the filler loading further increased to 20wt%, tensile strength declined slightly to 26.93MPa, although the elongation at break remained comparable to that of neat HDPE. This reduction may be associated with partial particle agglomeration or local matrix discontinuities at high filler contents, which weaken stress transfer efficiency. Similar effects have been reported in particulate-filled polymer systems when excessive filler loading promotes local agglomeration and weakens the continuity of the polymer matrix [10].

Overall, these results indicate that phosphorus slag powder can function effectively as an eco-friendly filler for HDPE composite applications. The developed materials satisfy the filler-related requirements for potential use in corrugated electrical conduit products, as specified in TCXDVN 272:2002. In addition to maintaining acceptable mechanical performance, the incorporation of phosphorus slag also contributes to reduced raw material cost and promotes the utilization of industrial by-products in plastic products.

3.2.3. Preparation of Putty powder (MTBT) using phosphorus slag powder

Wall putty powder (MTBT) formulations were developed and evaluated according to the technical requirements of TCVN 7239:2014. In conventional MTBT products, mineral fillers typically account for 55 - 80wt%, and therefore strongly influence the mechanical strength, workability, and cost efficiency of the final material [7]. Owing to the high whiteness, fineness, and improved chemical stability of the processed phosphorus slag, this material was investigated as a partial filler replacement in both exterior and interior MTBT formulations.

In the experimental formulations, DA 1120 redispersible polyvinyl alcohol (PVA) powder was used as the polymer binder. This material is commonly applied in putty and cement-based systems because of its contribution to water retention, adhesion, and mechanical performance. Its compatibility with inorganic fillers has also been reported in hybrid systems containing modified silicates and slag-derived materials.

Table 11. Formulations of some experimental MTBT samples (10 samples)

Form	Exterior MTBT					Interior MTBT				
Components	CT1	CT2	CT3	CT4	CT5	CT6	CT7	CT8	CT9	CT10
Black Cement	250	225	200	175	200	-	-	-	-	-
White Cement	-	-	-	-	-	360	324	288	252	200
Slag Powder	715	743	743	743	710	655	691	691	691	715
Lime	0	25	50	75	75	0	36	72	108	70
MK	28	0	0	0	2	36	0	0	0	2
Na_3PO_4	-	-	-	-	5	-	-	-	-	5
DA 1120	7	7	7	7	8	9	9	9	9	8
Total	1000	1000	1000	1000	1000	1000	1000	1000	1000	1000

A total of 10 MTBT formulations were prepared, including five exterior-grade samples (CT1-CT5) and five interior-grade samples (CT6-CT10), as listed in Table 11. The formulations were designed by varying the proportions of black cement, white cement, slag powder, lime, MK, Na_3PO_4, and DA1120 in order to identify compositions capable of meeting the technical criteria for putty applications.

After 7 days of curing, all formulations were evaluated in terms of fluidity, setting behavior, water retention, adhesion strength, and relative surface hardness. The detailed results are summarized in Table 12.

Table 12. Experimental Results According to TCVN 7239:2014

Sample, Mixture	Fluidity, cm	Setting time Start - Finish	Water retention, %	Adhesion strength, N/mm^2	Relative surface hardness
CT1	10	144 - 320	97	0.24	0.11
CT2	11	200 - 360	97.5	0.25	0.10
CT3	10.5	320 - 380	98.5	0.25	0.10
CT4	12	330 - 450	98.5	0.24	0.09
CT5	11.5	300 - 400	98.5	0.25	0.13
CT6	10	135 - 210	96.3	0.24	0.11
CT7	10.5	170 - 250	97.0	0.25	0.10
CT8	10.5	180 - 260	97.0	0.24	0.09
CT9	11.5	220 - 330	98.0	0.24	0.08
CT10	11	200 - 300	98.0	0.24	0.09
Note: Adhesion and Relative Surface Hardness Calculated After 7 Days					

Among the exterior-grade formulations, CT2, CT3, and CT5 exhibited the highest adhesion strength, reaching $0.25N/mm^2$, while CT1 and CT4 showed slightly lower values of approximately $0.24N/mm^2$. For the interior-grade formulations, CT7 reached the highest adhesion strength of $0.25N/mm^2$, whereas CT6, CT8, CT9, and CT10 showed values of approximately $0.24N/mm^2$. For the interior-grade formulations, CT10 was selected as the most representative composition due to its balanced technical performance, stable water retention, suitable workability, and optimized formulation design.

All measured adhesion strength values were close to or met the minimum requirement of $0.25N/mm^2$ specified in TCVN 7239:2014, indicating acceptable bonding performance of the developed putty systems. Notably, the white cement-based formulation (CT10) showed slightly more stable performance compared to the black cement-based series, which may be attributed to improved filler dispersion, matrix densification, and more favorable binder-filler interaction in the white-cement-based system.

In general, the formulated MTBT products showed high water retention (> 96%), suitable setting times (135 - 450 min), and acceptable relative surface hardness values (0.08 - 0.13). These results confirm that processed phosphorus slag can be effectively used as a major filler phase in MTBT systems without compromising the essential technical requirements. The favorable behavior may be related to the fine particle size, good packing ability, and calcium–silicate-rich character of the treated slag, which can contribute to matrix densification and improved filler distribution [5, 12].

In addition, the incorporation of residual phosphorus-bearing species within the calcium-silicate-rich slag matrix may reduce their mobility and reactivity, thereby contributing to the chemical stability of the final putty products [2, 3]. With further optimization, especially in terms of slag surface treatment and binder selection, phosphorus slag-based MTBT formulations may provide a promising route.

4. CONCLUSION

This study successfully processed and evaluated phosphorus slag powder for use in several filler-containing product systems, including wall putty, solvent-based and water-based paints, and HDPE-based thermoplastics. The results show that, after appropriate physical treatment, phosphorus slag powder can be incorporated over a relatively wide dosage range while maintaining product performance in accordance with relevant standards, including TCVN 7239:2014 for wall putty, TCVN 8789:2011 for protective coatings, and TCXDVN 272:2002 for plastic pipe-related materials.

The treatment route involving washing, impurity reduction, gravity separation, drying, and grinding effectively improved the quality of the slag powder and enabled its use as a substitute for conventional mineral fillers such as limestone powder. In the investigated formulations, the treated slag exhibited satisfactory compatibility and functional performance, confirming that phosphorus slag can serve as a feasible secondary mineral resource for non-structural and semi-structural applications.

From a broader perspective, the growing demand for mineral fillers in Vietnam's construction, coatings, and plastics industries is increasing pressure on non-renewable resources. In this context, the reuse of industrial by-products such as phosphorus slag offers a practical route for reducing waste-disposal pressure, lowering dependence on virgin minerals, and supporting more sustainable resource utilization. Overall, the results confirm that phosphorus slag from Tang Loong has clear potential for higher-value material applications beyond conventional low-value disposal pathways.

Conflict of interest

The authors agree that there is no conflict of interest with respect to the published results.

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