## THE POTENTIAL OF NICKEL SLAG WITH HUMIC SUBSTANCE ADDITION AS AMELIORATION MATERIALS ON GAJRUG RED-YELLOW PODZOLIC

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#### ABSTRACT

Nickel slag and humic substance have a strong potential for use as soil ameliorant in land remediation, owing to their chemical properties that can improve acidic soils. This study aimed to determine the effect of nickel slag and humic substance on the red-yellow podzolic soil. This greenhouse incubation study used a 2-factorial completely randomized design. The first Factor was nickel slag about 0, 8, 12, and 16 ton ha<sup>-1</sup> and the second Factor was humic substance about 0, 15, and 30 litre ha<sup>-1</sup>, with 3 replications. Statistical analysis of the data used analysis of variance, and whenever a significant difference was detected, further verification was tested using Duncan's Multiple Range Test at a level of significance ( $\alpha$ ) of 5%. The results showed that the application of nickel slag and humic substance can increase the values of pH, total-N, available-P, and exchangeable-Mg of the red-yellow podzolic soil, but also decreased exchangeable-H. The interactive effects of nickel slag and humic substance raise soil K-exchangeable only. The levels of heavy metals (Pb, Cd, and As) in the soil after the application of nickel slag and humic substance remain within normal limits (The normal limits for Pb, Cd, and As in soil are 2-200 mg kg<sup>-1</sup>, 0.1-7.0 mg kg<sup>-1</sup>, and 0.1-40 mg kg<sup>-1</sup>) that is, they are environmentally safe for use as a soil ameliorant.

Keywords: Heavy metal, humic substance, nickel slag, soil acidity

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## INTRODUCTION

Nickel processing produces by-products and solid residues, including slag that is formed when nickel ore is smelted and later goes through cooling. The rapid accumulation of nickel slag in slag dumps over time poses a serious problem attendant to industrial waste disposal. The slag requires large landfill sites; pollutes the environment, particularly when there are water bodies near the dump; and is a waste of resources which can otherwise, be recovered and recycled. There are potential some mining companies recover nickel slag and use it as a surfacing material for mining access roads. It is also being processed as an additive for abrasives for general industrial uses, and in ceramic and cement manufactures. There has been a number of studies on

utilization values of nickel slag. For instance,

the utilization potential of mining slag in agriculture however, these have been limited to steel slag. As an example, steel slag has been found to be a good material for liming acidic soils (Ali & Sedaghat Hoor, 2007; Ning, Liang, Liu, Xiao, & Duan, 2016); as silicon (Si) fertilizer for rice paddy and sugar field (Kato & Owa,

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1997); for improvement of the chemical properties of soil; and for boosting the growth and yield of rice in paddy field and in peatland (Ulfah, 2014).

Unfortunately, the utilization of nickel slag in agricultural production is constrained by the fact that it falls under category Hazardous and Toxic Substances (in Bahasa Indonesia Limbah Bahaya dan Beracun = B3) waste material, the handling of which must strictly follow existing government regulations. In particular, PP No. 101/2014 ir: Hazardous and Toxic Wastes, requires prior Toxicity Leaching Procedure (TCLP) Characteristic testing. Interestingly, previous TCLP tests for nickel slag have yielded results that fall belowset quality specifications. In other words, nickel slag is actually "not hazardous" and therefore, should not be classified as a B3 waste. Hence, nickel slag should be re-classified and promoted to be utilized, instead of simply left lying idle and accumulating in slag dumps. For instance, nickel slag can be used in agricultural cultivation, as it contains useful chemical elements such as K, Ca, Mg, Na, and Si which are suitable as liming agents for soil amelioration.

It is interesting to note that the chemical composition of nickel slag is quite similar to that of steel slag. It thus, follows that nickel slag can be as suitable as steel slag for soil amelioration purposes. There is. however another major constraint in that the solubility of nickel slag elements is rather poor due to its inherently hard physical nature. To remedy this limitation, a suitable chemical compound must be added to the nickel slag in order to improve its element solubility. One feasible material that can fill this need is a humic substance, as it contains organic acids that can effectively dissolve minerals through the chemical reaction (e.g. hydrolysis, acidolysis, complexolysis). This is made possible by the high negative load of humic substances, which

indirectly facilitates chelation of cations and increase cation exchange capacity in the soil. In addition, the humic substance can interact with metal ions, oxides, hydroxides, and other contaminant substances (Schnitzer & Khan, 1978). (Ahmad, 2011) also reported that humic substance serves as a good extractor for igneous rock, and in the release of nutrient elements. Furthermore, it renders an ameliorating influence on the physical, chemical, and biological aspects of soil quality.

The utilization potential of nickel slag mixed with humic substance as ameliorant material can be applied to red-yellow podzolic (RYP) soil, which is characteristically acidic. Covering a wide expanse in Indonesia (nearly 25% of the total land area of the country), RYP soil has tremendous utilization potential for dryland agricultural development. Nevertheless, there is a need to address the problem of very low nutrient content in RYP soil, as well as its high Aluminium saturation that can bring about Al<sup>3+</sup> poisoning in plants (Havlin & Tisdale, 2005).

In view of the foregoing, this study intended to analyze the efficacy of nickel slag and humic substance in improving the chemical characteristics of Red Yellow Podzolic soil, particularly its pH, macronutrient element content, bases content, exchangeable acids, cation exchange capacity (CEC), as well as its concentration of heavy metals (Pb, Cd, As).

## MATERIALS AND METHODS

Samples of red-yellow podzolic (RYP) soil was collected from Gajrug village, Cipanas subregency, Banten province in West Java, Indonesia. The chemical properties of the Gajrug RYP soil are shown in Table 1. The humic substance was received from the Department of Soil Science and Land Resources, Bogor Agricultural University. Table 2 summarizes the results of the chemical analysis of the humic substance. Nickel slag was obtained from a nickel mine in South Sulawesi, Indonesia. Data on the properties of the nickel slag are shown in Table 3.

This study involved an incubation experiment using a 2-factorial Completely Randomized Design (CRD). The first Factor was nickel slag (T) about T0, T1, T2, and T3 corresponding to 0, 8, 12, and 16 ton ha<sup>-1</sup> and the second Factor was humic substance about H0, H1, and H2, corresponding to 0, 15, and 30 litre ha<sup>-1</sup>, respectively, in the field, with 3 replications, or a total of 36 experimental units.

#### **Incubation Media Trial**

The RYP soil samples used as incubation media were obtained from several sampling points in the field at a depth of 0-20 cm and composited. Then, they were air-dried for 2 weeks sifted using a 5-mm sieve, and 36 airdried samples, each weighing 0.5 kg, were prepared. The soil samples were placed inside plastic glasses for the incubation trial with the ameliorant material, at the specified dosage levels. The nickel slag was ground using a grinding stone and sifted with a 65 mesh. The nickel slag that passed the sieve was weighed following the set dosage levels of each treatment, and placed inside the incubation media. Afterward, the incubation media were applied with basic fertilizer at dosages of 0.14 g pot<sup>-1</sup> (corresponding to 300 kg ha<sup>-1</sup> urea), 0,06 g pot<sup>-1</sup> (corresponding to 100 kg ha<sup>-1</sup> SP-36), and 0.04 g pot<sup>-1</sup> (corresponding to 100 kg ha<sup>-1</sup> KCI).

The humic substance was first diluted with 100 times distilled water. Then, the diluted humic substance was added into the plastic glasses that already contained the prepared levels of nickel slag. Afterward, the plastic glasses were put inside the greenhouse, with the experimental RYP soil being maintained at field capacity for the whole incubation period. After 6 weeks, the RYP soil samples were collected for laboratory analysis.

Parameter	Unit	Determination Method/Measuring Device	Value
рН	-	pH meter	4.03
Organic-C	%	Walkley & Black	0.07
Total-N	%	Kjeldahl	0.08
Available-P	Ppm	Bray 1	7.92
exchangeable-K	cmol (+) 100g <sup>-1</sup>	NH₄OA <sub>c</sub> pH 7	2.30
Exchangeable-Ca	cmol (+) 100g <sup>-1</sup>	NH <sub>4</sub> OA <sub>c</sub> pH 8	13.07
Exchangeable-Na	cmol (+) 100g <sup>-1</sup>	NH₄OA <sub>c</sub> pH 9	0.84
Exchangeable-Mg	cmol (+) 100g <sup>-1</sup>	NH₄OA <sub>C</sub> pH 10	0.54
CEC	cmol (+) 100g <sup>-1</sup>	NH₄OA <sub>C</sub> pH 11	20.31
Exchangeable-Al	cmol (+) 100g <sup>-1</sup>	KCI 1 N	30.12
Exchangeable-H	cmol (+) 100g <sup>-1</sup>	KCI 1 N	4.18
Heavy metal:			
Pb	mg L <sup>-1</sup>	AAS (Atomic Absorption Spectroscopy)	<0.038
Cd	mg L <sup>-1</sup>	AAS (Atomic Absorption Spectroscopy)	0.025
AS	mg L⁻¹	AAS (Atomic Absorption Spectroscopy)	0.012

#### **Table 1.** Chemical Properties of the Gajrug RYP Soil before Treatment

Note: results of the pre-research analysis

Type of Analysis	Method/Instrument	Unit	Value	Remarks
рН	pH meter	-	8.71	In the concentrated liquid form
Electrical	EC meter	mS cm⁻¹	13.89	In the concentrated liquid form
Conductivity				
Carbon content	CNS ( <i>C, N and S</i>	%	41.77 *	In solid form
	analyzer)			
Nitrogen content	CNS ( <i>C, N and S</i>	%	1.17 *	In solid form
	analyzer)			
Ash content	Gravimetric	%	2.96 *	In solid form

Table 2. Properties of the Humic substance

\* Calculated based on the level of density (7.74%) Source: (Oklima, 2014)

#### Table 3. Properties of the Nickel Slag

Compound	Total (%)	Determination Method/Measuring Device
SiO <sub>2</sub>	45.30	Gravimetric
$AI_2O_3$	2.53	AAS (Atomic Absorption Spectroscopy)
$Fe_2O_3$	21.30	AAS (Atomic Absorption Spectroscopy)
K <sub>2</sub> O	0.17	AAS (Atomic Absorption Spectroscopy)
Na <sub>2</sub> O	0.72	AAS (Atomic Absorption Spectroscopy)
CaO	1.00	AAS (Atomic Absorption Spectroscopy)
MgO	23.20	AAS (Atomic Absorption Spectroscopy)
MnO	0.63	AAS (Atomic Absorption Spectroscopy)
TiO <sub>2</sub>	0.27	Spectrophotometer
$P_2O_5$	0.21	Spectrophotometer

Note: results of the pre-research analysis

The chemical parameters of the incubation media that were evaluated in this study included pH H<sub>2</sub>O (using a pH meter); total-N (by means of *Kjeldahl method*); available-P (using Bray 1 *method*); exchangeable-K, exchangeable-Ca, exchangeable-Na, exchangeable-Mg, and CEC (with NH<sub>4</sub>OA<sub>c</sub> pH 7.0); exchangeable-Al and exchangeable-H (using KCl 1N); together with heavy metal content (Pb, Cd, and As) of the dissolved soil (using diethylene triamine penta acetic acid (DTPA) extraction). Statistical analysis of the data was done using Analysis of Variance (ANOVA) at ( $\alpha$  =) 0.05 level of significance, and when a significant difference between treatments was observed, further statistical verification was carried out with Duncan Multiple Range Test (DMRT), also at  $\alpha$  = 0.05 level.

## Chemical Characteristics of the RYP Soil, Humic substance, and Nickel Slag

The chemical properties of the Gajrug RYP soil, humic substance, and nickel slag are presented in Table 1, Table 2, and Table 3, respectively.

## RESULTS AND DISCUSSION Soil Reaction

Table 4 showed the data on the Gajrug RYP soil after treatments. Nickel slag caused a significant increase in the RYP soil pH. As depicted in Table 4, the treatment of nickel slag at level 8 ton ha<sup>-1</sup>, even without humic substance (T1H0), produced the greatest effect in raising soil pH (from 4.03 to 4.63). The increase of pH value was due to the presence of CaO and MgO (Table 3) which are released from nickel slag into the soil solution. Both CaO and MgO compounds found in the nickel slag reacted with  $H_2O$  forming  $Ca(OH)_2$  and  $Mg(OH)_2$  which disintegrated and became  $Ca^{2+}$  +  $2OH^-$  and  $Mg^{2+}$  +  $2OH^-$ . The  $OH^-$ ions bonded with  $H^+$  to become  $H_2O$ , therefore,  $H^+$  ions that caused acidity decreases.

The nickel slag neutralizing capacity is not as high as the steel slag since the CaO in steel slag is higher (53.36%) (Gultom, 2012) than in nickel slag. In addition, based on the acid neutralization capacity (ANC) analysis of the nickel slag obtained value of 31.9%, which was far lower than that of steel slag (66.08%), according to (Utami, 2012), and dolomite (109%) when calculated based on CaCO<sub>3</sub> equivalence. This implies that in neutralizing soil acidity, the nickel slag can neutralize only 31.9% compared to CaCO<sub>3</sub> (100%). Although its acid neutralization capacity (ANC) is relatively low, nickel slag renders a liming effect on the Gajrug RYP soil.

#### **Total Nitrogen**

**Table 4.** Effect of the application of nickel slagand humic substance on soil pH

Humic substance			
H0	H1	H2	
4.51 ab	4.55 a	4.58 a	
4.63 a	4.52 a	4.60 a	
4.53 b	4.47 a	4.47 a	
4.52 b	4.53 a	4.53 a	
	Hum H0 4.51 ab 4.63 a 4.53 b 4.52 b	Humic substan           H0         H1           4.51 ab         4.55 a           4.63 a         4.52 a           4.53 b         4.47 a           4.52 b         4.53 a	

Numbers in the same column that are followed by similar letters denote not-significant difference at 5% significance level using Duncan's Multiple Range Test (DMRT).

**Table 5.** Effect of the application of nickel slagand humic substance on soil total-N (%)

	Humic substance			
NICKEI Slag	HO	H1	H2	
Т0	0.11 b	0.10 a	0.11 a	
T1	0.11 b	0.10 a	0.12 a	
T2	0.12 a	0.14 a	0.14 a	
Т3	0.11 b	0.10 a	0.09 a	

Numbers in the same column followed by similar letters denote not-significant difference at 5% level of significance, using Duncan's Multiple Range Test (DMRT).

The effects of adding nickel slag and humic substance on the total-N content of the Gajrug RYP soil are shown in Table 5. The results of the various statistical tests suggested that only the application of nickel slag produced a measurable effect on soil total-N.

Statistical analysis also established that the application of nickel slag at a level of 12 ton ha<sup>-1</sup> (T2) produced the biggest effect in raising soil total-N (from 0.08% to 0.12%). According to (Munawar, 2011), when the soil is acidic, microbial activity is impeded, as nitrogen-fixing bacteria in the air are generally sensitive to a low pH level leading to a reduction in N intake. While humic substance, both at the level of H1 and H2 are not a significant reaction because the suspected dose is still too low to use. this is due to the humat substance are easily dissolve so that the rate of nitrogen release process in the soil is more quickly disappear before being bound by the soil material.

**Table 6**. Effect of the application of nickel slag and humic substance on soil available-P (ppm  $P_2O_5$ )

2 = 51				
Nickel	Humic substance			
Slag	H0	H1	H2	
Т0	11.17 b	13.90 a	11.65 a	
T1	11.29 ab	9.51 a	10.46 a	
T2	19.25 a	14.62 a	12.60 a	
Т3	12.95 ab	11.88 a	12.84 a	

Numbers in the same column that are followed by similar letters denote not-significant difference at 5% level of significance, using Duncan's Multiple Range Test (DMRT).

 Table 7. Effect of the application of nickel slag

 and humic substance on soil exchangeable-H

 (cmol (+) 100g<sup>-1</sup>)

	· · ·	0,			
	Nickel	Hu	Humic substance		
	Slag	HO	H1	H2	
_	Т0	2.48 a	4.17 b	3.44 a	
	T1	3.49 a	3.28 a	2.86 a	
	T2	0.81 b	3.02 a	1.84 a	
	Т3	2.76 ab	4.16 a	1.59 a	

Numbers in the same column that are followed by similar letters denote not-significant difference at 5% level of significance, using Duncan's Multiple Range Test (DMRT).

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#### **Available Phosphor**

Table 6 summarizes the effect of the application of nickel slag and humic substance on available-P in the Gajrug RYP soil. Statistical analysis revealed that only the application of nickel slag produced a significant effect on soil available-P.

When subjected to the statistical test, the data showed that the application of nickel slag significantly increased available-P of the Gajrug RYP soil (Table 6), with treatment T2 (12 ton ha<sup>-1</sup>) producing the greatest increase (from 7.92 ppm to 19.25 ppm). This increase in the level of soil available-P could have been due to the P<sub>2</sub>O<sub>5</sub> that was released from the nickel slag (Table 3) into the RYP soil solution. In addition, the SiO<sub>2</sub>contained in the nickel slag was likewise released into soil solution hence, also contributed to the increase in the level of soil available-P (Myhr & Erstad, 1996). The Si content of the nickel slag (Tabel 3) also reduced P fixation by Al and Fe through ligand exchange, such that the  $SiO_2$  in the nickel slag was hydrolyzed and formed SiO<sub>4</sub><sup>4-</sup> anions that replaced PO<sub>4</sub><sup>3-</sup>. This resulted in the release of P in the nickel slag into the soil solution.

#### Exchangeable-H

The effect of the application of nickel slag and humic substance on the level of exchangeable-H of the Gajrug RYP soil can be seen in Table 7. The application of nickel slag and humic substance generally lowered the level of exchangeable-H in the RYP soil.

The application of 12 ton ha<sup>-1</sup> nickel slag without humic substance (T2H0), produced the highest reduction (from 2.48 cmol(+)100g<sup>-1</sup> to 0.81 cmol(+)100g<sup>-1</sup>. Allorerung (1988) found that the Si contained in the steel slag indirectly influenced the reduction in exchangeable-Al and exchangeable-H inRYP soil, as part of the silicates released (by the steel slag) into the soil solution reacted or settled with Al and Fe hydroxide. The same thing likely happened with the nickel slag considering its chemical composition (45.30%) is also  $SiO_2$  (Table 3).

# Exchangeable Cation (exchangeable-K, exchangeable-Mg)

The application of nickel slag and humic substance significantly increased exchangeable-K of the RYP soil and the single application of the humic substance (H1TO) resulted in the highest level of exchangeable-K (1.08 cmol(+) 100 g<sup>-1</sup>.

As shown in Table 8, the addition of nickel slag into the Gajrug RYP soil significantly raised the level of soil exchangeable-Mg, notably rising with increasing dosage.



Numbers in the bar graph that are followed by similar letters denote not-significant difference at 5% level of significance, using Duncan's Multiple Range Test (DMRT).

**Figure 1.** Graph showing the effect of the application of nickel slag and humic substance on exchangeable-K of the RYP soil

 Table 8. Effect of the application of nickel slag

 and humic substance on soil exchangeable-Mg

 (cmol(+)100g<sup>-1</sup>)

· _	· · · ·	, ,		
	Nickel	Humic substance		
_	Slag	HO	H1	H2
	Т0	14.80 c	15.56 a	14.00 a
	T1	18.14 b	17.66 a	19.04 a
	T2	18.93 b	19.53 a	19.04 a
	Т3	21.33 a	21.64 a	21.77 a

Numbers in the same column that are followed by similar letters denote not-significant difference at 5% significance level, using Duncan's Multiple Range Test (DMRT).

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**Table 9.** Effect of the application of nickel slag and humic substance on the rate of some heavy metals in the soil (ppm)

Tractment	Heavy Metal			
freatment	Pb	Cd	As	
тоно	0.0126	0.0005	0.2169	
T0H1	0.0075	0.0019	0.2467	
T0H2	0.0187	0.0008	0.2030	
T1H0	0.0190	0.0005	0.1925	
T1H1	0.0081	0.0010	0.1577	
T1H2	0.0090	nm	0.2104	
T2H0	0.0215	0.0007	0.2055	
T2H1	0.0081	0.0007	0.2064	
T2H2	0.0191	0.0007	0.2745	
T3H0	0.0140	0.0012	0.2424	
T3H1	0.0256	0.0015	0.2497	
T3H2	0.0199	0.0012	0.2190	

note: nm – not measured

The highest nickel slag level of 16 ton ha<sup>-1</sup> (T3) in Table 8 produced the greatest increase in exchangeable-Mg (21.33 cmol(+)100g<sup>-1</sup>). This could be attributable to the fact that nickel slag possesses a high Mg level, as manifested by its high MgO content (Table 3).

## Heavy Metals in the Soil

The results of the humic substance (Table 2) and the nickel slag (Table 3) showed the absence of Pb, Cd or As in humic substance and nickel slag. However, in the TOHO (treatment soil without humic substance or nickel slag) it was found Pb, Cd and As. This indicates that the three heavy metals are already present in the soil (Table 1), but these are still in normal environmental standard. The critical limits for Pb, Cd, and As in soil are 100-400 mg kg<sup>-1</sup>, 3-8 mg kg<sup>-1</sup>, and 20-50 mg kg<sup>-1</sup>, respectively (Alloway, 1995). While the normal limits for PB, Cd, and As in soil are 2-200 mg kg<sup>-1</sup>, 0.1-7.0 mg kg<sup>-1</sup>, and 0.1-40 mg kg<sup>-1</sup> (Barchia, 2009).

The application of humic substance and nickel slag were tending to decrease the content of Pb and As in the soil. It can be seen in Table 9 that the levels of Pb, Cd, and As that were extracted using DTPA were lower than those obtained by Alloway (1995). The decrease in the heavy metal content of Pb and As is thought to be due to a reaction between heavy metals with a humic material forming a complex bond.

## CONCLUSION

Based on the statistical analysis of the data obtained in this study, it is clear that the application of nickel slag into the Gajrug redyellow podzolic soil can increase significantly the levels of soil pH, total-N, available-P, exchangeable-Mg. At the same time, the treatment can decrease exchangeable-H in the soil. The interaction effect of adding nickel slag and humic substance application into the RYP soil can increase exchangeable-K only. The application of nickel slag and humic substance into the Gajrug RYP soil tended to decrease heavy metals, notably Pb, Cd and As which implies they pose no environmental hazard, and can safely be used as a soil ameliorant. The concentration of the humic substance (both H1 and H2) should be increased so that the reaction in the soil is more optimal, especially in helping to dissolve the element of nickel slag so that the Gajrug red-yellow podzolic soil quality is better.

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