Evaluating the Optical Properties of Philippine Rice Hull Ash as a Potential Alternative to Calcium Carbonate Filler for Paints

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Abstract- The exploration of silica-rich agricultural wastes can be attributed to the constant pursuit of adding sustainable materials in the coatings industry. Inarguably, rice hull ash or RHA is one of the most studied agricultural wastes due to its high silica content and has been the subject of numerous research investigations. Owing to it excellent properties, RHA is used as pozzolan in the construction industry, as a filler, additive, oil adsorbent, and suspending agent for porcelain enamels. This research study attempts to explore and evaluate the Philippine rice hull ash as potential alternative to calcium carbonate fillers in term of its optical properties. A series of processes and treatments, which include milling and firing, were used to yield a white RHA product resembling a calcium carbonate. The resulting RHA was characterized using analytical instruments and its optical properties were obtained using a colorimeter for comparison with the commercially available calcium carbonate fillers. The optical properties of the processed RHA used in this study generated a brightness level, L*, that is within and is higher than the brightness level of the calcium carbonate populated in this study. Hence, in terms of optical properties, RHA can be processed to be used as an alternative mineral filler to paints. However, this research is delimited by the chemical and functional characteristics of the rice hull ash when blended and formulated with a paint or coating. A separate study must be undertaken to evaluate the viability of rice hull ash as extender to paints.

Indexed Terms- Alternative Filler, Optical Properties, Paint Fillers, Rice Hull Ash

I. INTRODUCTION

In the recent years, sustainability has become increasingly important as ancillary to a nation's social, economic, and environmental growth. Organizations from different industries are making conscious effort to incorporate sustainability within their companies. In the paints and coatings industry, there has been a trend for going "green" by replacing non-renewable raw materials with sustainable ones, including fillers. Fillers are solid particles used in paints and coatings system to occupy a large volume of the formulation, thereby, reducing its cost. Fillers also improve paints and coatings by conferring some properties to it such as abrasion and weathering resistance (Special Chem, n.d.). Some of the most used paint fillers are calcium carbonate, kaolin, and silica flours (Addullah, M., et. al., 2021). Others include talc, wollastonite, and barium sulphate.

The quest for increasing the sustainable content of paints has led to the exploration of silica-rich agricultural wastes which include rice husk as an extender or filler. Rice hull or rice husk is inarguably one of the most common sources of biomass fuel. Combustion of rice hulls in powerplants produce waste in form of rice husk ash, also rice hull ash (RHA) which is known to possess high silica content and pozzolanic properties. Beneficiation of RHA does not only add value to the sustainability of paints but also provides solution to the bulk of agricultural waste disposed in landfills and open fields which pose environmental concern and potential human related problems, owing to the low bulk density of RHA (Pode, R., 2016).

As one of the most at risk countries from the climate crisis according to the Institute for Economics and Peace (Amnesty International, 2021), the Philippines is actively fighting climate change by committing to a projected greenhouse gas emissions reduction and avoidance of 75% by 2030 (USAID, n.d.). And with a huge dependence on the agriculture, the country produces substantial amount of agricultural waste, the most common of which are, rice hull, bagasse, coconut shell husk and coconut coir (ASEAN Briefing, 2017). Most of these agricultural wastes are used as biofuels in biomass powerplants as in the case with rice husk. RHA is used in various applications such as as a filler, additive, abrasive agent, oil adsorbent, sweeping component, and suspending agent in porcelain enamels. It is also used as a pozzolan in the building sector (Singh, B., 2018). The high silica content of RHA has been investigated through various research studies to potentially replace portion of some of the common filler in paints but with no mention of the optical properties.

This study attempts to evaluate the optical properties of Philippine rice husk ash as a potential substitute to calcium carbonate paint filler.

II. LITERATURE REVIEW

- Study and Use of Rice Husk Ash as a Source of Aluminosilicate in Refractory Coating

Using rice husk ash (RHA) as an aluminosilicate source in fire-resistant coatings has the potential to lower pollution levels in the environment and create industrial wealth from agricultural waste. Increasing RHA contents showed an increase in the total and open porosities and rough surfaces which directly influence the formation of the intumescent char layer since the number of pores plays an important role on the coating surface. Given its well-known high silica concentration, rice husk ash (RHA) presents a significant opportunity as a substitute for commercial paint additives or fillers. Nowadays, the most extensively used fillers in the paint industry are calcium carbonate, kaolin, and silica flour since they are naturally occurring and have excellent crystalline silica. According to recent research, RHA can potentially be utilized as a filler. The most significant benefit is that it can enhance some of the mechanical qualities of epoxy paints. It is also affordable and renewable. (Addullah, M., et. al., 2021)

- Epoxy Coatings Fillers on the Rice Husk Base

For the manufacturing of epoxy polymer fillers, especially those based on synthetic wollastonite, rice husk and ash are highly desirable. Because rice husk contains a significant amount of amorphous silica that is chemically active, it may be used as an effective filler for linear and cross-linked polymers because it has a catalytic effect on the amine-induced hardening of epoxy oligomers, which lowers the degree of cross-linking in epoxy polymers (Gotlib, E. et. al., 2019).

- Pigment Extender Properties of Rice Husk Ash in Emulsion Paint

Rice husk ash (RHA) can be used as a pigment extender in emulsion paints, according to this study. The outcomes also showed that the RHA-filled emulsion paints performed well and had application qualities that met the standards for premium paints. RHA is a cheap, abundant, and good renewable extender for emulsion paints (Dilim, C., 2007).

- Rice Husk ash as Flatting Extender in Cellulose Matt Paint

Rice husk ash is a cheap, abundant, and renewable resource that can be used for a variety of industrial purposes, including the formulation of surface coatings, due to its high silica concentration. To determine if rice husk ash (RHA) may be used as a flatting extender in cellulose matt paint, this study compared the flatting effects of RHA with those of two commercial extenders: silica flour and fumed silica, which is the usual flatting agent (Igwebike-Ossi, C.D., 2014).

- Flatting Effect of Rice Husk Ash in Red Oxide Primer in Comparison with Calcium Carbonate

A growing interest in agricultural waste extenders with possible usage in paints and other surface coatings has been sparked by the world's slow shift away from the use of non-renewable (fossil-based) resources and toward renewable resources. In recent times, there has been an investigation into the suitability of RHA as an extender in the surface coatings business for matte wood varnish, cellulose matt paint, and textured emulsion paints (Igwebike-Ossi, C. and Elom, N., 2016).

- The use of rice husk ash as reactive filler in ultra-high-performance concrete

RHA's chemical composition varies according to the conditions of combustion, especially at high temperature. Only crystalline silica is left over after burning at temperatures exceeding 700°C which is utilized in the steel and ceramic industries. It has been suggested that RHA should be burned at low temperatures because crystalline silica can be harmful to human health when exposed to air since it can cause silicosis. Amorphous silica, which is utilized as a filler in paint or rubber as well as an additional cementitious material (SCM) in building materials, is created by burning rice husks below 700°C (Kang, H. et. al., 2019).

III. METHODOLOGY

The research design that will be implemented to evaluate the optical properties of Philippine rice hull ash as a potential substitute to calcium carbonate paint filler is an applied research design which seeks to provide practical application and innovative solution to societal problems. In this case, the study intends to identify the potential of rice hull ash as paint additive in terms of optical properties which makes it an additional option for the encompassing utilization of this agricultural waste. The study will also employ an experimental research design. The research study will be conducted at a laboratory located in Pampanga, Philippines. Coincidentally, Pampanga is part of the Central Luzon region, the Rice Granary of the Philippines, due to its abundance of plains and farmlands (NEDA, 2021). Central Luzon was the top producer of rice in 2023, producing 18% or 3.6 million metric tons out of the more than 20 million MT of palay harvested (Daily Tribune Lifestyle, 2024). Sample rice hull ash, on the other hand, was obtained from Bulacan, also a province in the region. The laboratory is equipped with the necessary equipment and analytical tools to meet the objectives of this research study.

The researcher shall use digital weighing scale, Konica Minolta colorimeter, an XRF device, XRD equipment, Malvern particle sizer, and Microsoft Excel as data collecting instrument in this research paper. The colorimeter shall be used to obtain the CIELAB colour space optical properties, that is, L*a*b. Meanwhile, the XRF device shall be utilized to get the elemental composition of the rice hull ash while particle sizer will be used to get the size fraction distribution.

Descriptive analysis shall be used to evaluate, analyse, and interpret the result of the experimentation whether the optical properties of Philippine rice hull ash is a potential substitute to calcium carbonate paint filler. The descriptive research design allows facts to be described by establishing relationship between variables. In this case, the study aims to identify the suitability of the optical properties of the Philippine rice hull ash in terms of CIELAB colour space in respect to calcium carbonate paint filler.

IV. RESULTS AND DISCUSSIONS

- Characterization of the Rice Husk Ash

Rich husk ash is known to contain high amounts of silica, but the chemical composition of RHA varies according to the combustion conditions, processes involved, and other factors. Firing RHA at temperature below 700C is known to produce amorphous silica which is used as supplementary cementitious material in the construction industry and as filler for rubbers and paints (Kang, H. et. al., 2019). In this study, the elemental oxide of the RHA used was obtained using an X-ray Fluorescence. This determines the purity and the amount of silica, and other minerals present in the agricultural waste. Table 1 illustrates the elemental oxide composition of the processed RHA.

Oxide (wt%)	Rice Husk Ash	Oxide (wt%)	Rice Husk Ash
Na_2O	0.06	Fe_2O_3	0.05
MgO	0.44	NiO	<0.01
Al_2O_3	0.17	CuO	<0.01
SiO_2	**95.24**	ZnO	<0.01
P_2O_5	0.54	SrO	0.01
SO_3	<0.01	ZrO_2	<0.01
K_2O	1.72	BaO	<0.01
CaO	0.56	HfO_2	<0.01
TiO_2	<0.01	PbO	<0.01
V_2O_5	<0.01	L.O.I.	0.3
Cr_2O_3	<0.01	TOTAL	99.2
Mn_3O_4	0.11	Ash Content	88.47

Table 1 XRF Analysis of the RHA subject

Consistent to the general silica content of RHA which is 94-96% (Putranto, A., et. al, 2021), the subject RHA of this research study contained 95.24% silica, with 1.72% potassium oxide, and 0.56% calcium oxide. Loss of ignition is 0.3%. The remaining are below 0.5% of other elemental oxides. The high silica content in the RHA presents opportunity for further processing into potential filler substitute for paints.

- Particle sizing

Raw RHA is wet micronized through a proprietary formulation using planetary ball mills for one hour at a specific speed. This process breaks down the RHA's particles' sizes into finer fractions to resemble or at least be comparable with calcium carbonate. The particle size distribution was determined using laser diffraction via Mastersizer 2000 from Malvern at 1725 rpm. The average RHA particle size at D50 was determined to be 3.497 micron while D10 and D90 were 1.604 and 6.115, respectively. Table 2 shows the particle size distribution of the RHA after milling.

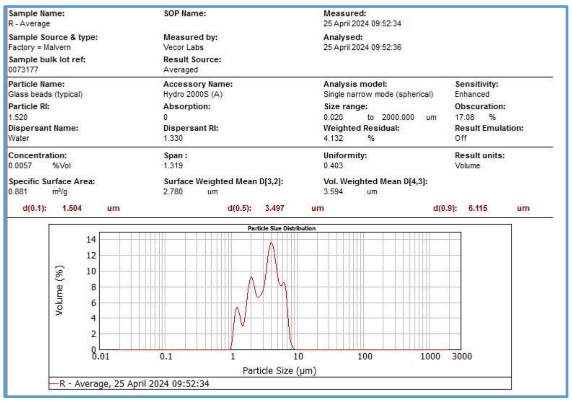

Table 2 Particle size distribution of the milled RHA

- Firing

Using a top load kiln, micronized RHA was rinsed, dried, and fired at a specific temperature and soaking period. RHA calcined at 700C and below produces amorphous silica while when calcined at 700C and up, crystalline silica is produced (Kaleli, J., et.al., 2020). In this research study, the RHA was calcined and fired at temperatures below 700C and above 1000C, respectively. Resulting RHA from <700C yielded dark appearance while at >1000C, it yielded a white product. Moreover, at temperature above 1000C, it can be seen from the X-ray diffraction (XRD) at Figure 1 that the silica content from the RHA showed crystalline phase, which is consistent to previous studies. The resulting physical images are shown in Figure 2.

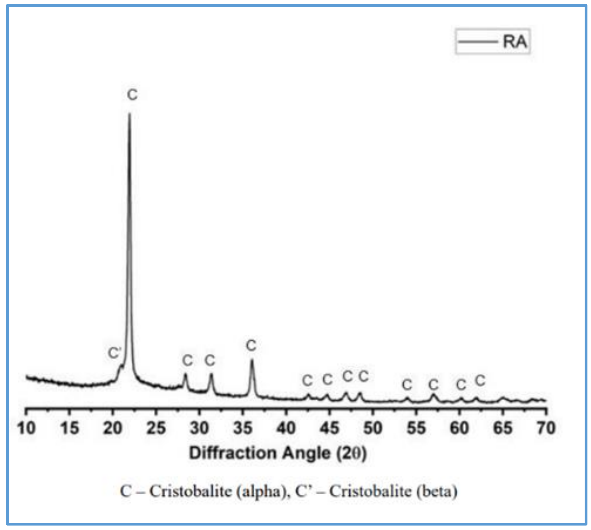

Figure 1 XRD of the RHA fired at temperature above 1000C.

Figure 2 Fired RHA at <700C (left) and fired RHA at >1000C (right)

- Optical Properties Color Space CIELAB RHA

Konica Minolta CR-20 colorimeter was used to obtain the color space CIELAB of the resulting processed rice hull ash. The acronym for the International Commission on Illumination, Commission Internationale de l'Eclairage in French, is where the name "CIE" in CIELAB originates. L*a*b* represents color space used to measure objective color and calculate color differences. L* represents lightness from black to white on a scale of zero to 100, while a* and b* represent chromaticity with no specific numeric limits. It is known that negative a* is associated with green, positive a* with red, negative b* with blue, and positive b* with yellow (Patel, J.B., and Anand, R.S., 2012). One standard of L*a*b* measurement is based on ASTM E 1164, Standard Practice for Obtaining Spectrometric Data for Object-Color Evaluation. Essentially, the higher the value for L*, the brighter it is. For this study, only the L*a*b* of the RHA sample fired at >1000C was obtained (see Table 3) since the other sample demonstrated an incomplete combustion of carbon resulting to black color appearance.

Material	Temp	L	a	b
RHA	>1000C	97.40	1.00	2.00

Table 3 shows the CIELAB values of the processed RHA.

- Comparison of CaCO3 and RHA

The values of L* of the RHA and several commercially available CaCO3 were tabulated and analyzed using Microsoft Excel. The study yielded a close optical property comparison for these two materials. Note that only the brightness level, L*, is obtained for comparison since this is the only optical parameter available in every calcium carbonate technical datasheet. A comparison of brightness levels is shown in Table 4.

Brightness level, L*, of the commercially available and known calcium carbonate paint fillers were obtained from some of the major players in the calcium carbonate industry which are Omya, Imerys, Huber, and Sibelco. The tabulation of the L* values of these calcium carbonates against the processed rice hull ash revealed that the latter's brightness level is within and above the values of some of these paint fillers in the market. The processed RHA's L* value is highest amongst the samples at 97.4 followed by Portafill H10 at 97.2, Hubercarb G35T (95), Omyacarb 5-MH (94.5), Calcigloss – IP (94), and Imercarb 7 with 92 L* value. Therefore, in terms of optical properties, it can be inferred that RHA is comparable and can be used as an alternative to calcium carbonate as paint filler when we talk about the brightness level of the optical properties.

Brand	Brightness, L*	Type	Manufacturer
RHA	97.40	Rice hull ash	-
Hubercarb® G35T	95	Ultrafine ground calcium carbonate	Huber
Imercarb 7	92	calcium carbonate	Imerys
OMYACARB® 5 - MH	94.5	dispersible calcium carbonate	Omya
Calcigloss - IP	94	very fine calcium carbonate	Omya
Portafill H10	97.2	partially hydrated magnesium-calcium carbonate	Sibelco

Table 4 Comparison of brightness level of the processed RHA versus commercially available CaCO3 paint fillers

V. SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

- SUMMARY OF FINDINGS

In this study, the Philippine rice hull ash obtained from Bulacan province was subjected to different methods and procedures to evaluate whether its optical properties, when processed, can be an alternative to mineral paint filler such as calcium carbonate. The RHA was characterized using an XRF, milled to a micron level, then assessed via particle sizer equipment. It was fired in two different temperatures, <700C and >1000C. Initially, the RHA was calcined at <700C, but incomplete combustion led to its dark appearance which makes optical measurement of whiteness or brightness inapplicable. It was then fired at >1000C at a specific duration which yielded a white RHA output. The CIELAB values of the processed white RHA were obtained using a colorimeter and were compared with other commercially available calcium carbonate used as paint fillers. The result yielded a comparable L* value or brightness level of the RHA (97.4) with the calcium carbonates (92 to 97.2 brightness level). Hence, in terms of optical properties alone, RHA can be used as a substitute for commercial calcium carbonate fillers. But in terms of its compatibility with pain formulation, this should be a subject for another comprehensive testing and research.

- CONCLUSION

The result of this study has allowed the further exploration of the beneficiation of rice husk ash as potential substitute of mineral paint filler calcium carbonate in terms of its optical properties. The study has demonstrated that in terms of optical properties through the numerical value of the CIELAB, processed RHA is comparable with commercially available calcium carbonate used as paint fillers.

- RECOMMENDATIONS

This research study is aimed to give light whether rice hull ash can be used as a potential substitute to mineral paint filler like calcium carbonate in terms of their optical properties through the CIELAB color space. In this essence, the study is delimited only to this comparison. Should other researcher wish to find out the effect of replacing the calcium carbonate with this processed rice hull ash, whether by full or partial replacement, shall be subject to another comprehensive research study which involves paint formulation and other related testing. Also, previous studies state that firing the RHA at >1000C is suitable for ceramic applications whereas firing the material at <700C makes it suitable for filler applications. However, calcining at this low temperature yielded black color in this study. Therefore, it is also recommended to reconcile producing a white RHA product at temperature below 700C where silica is amorphous, intended as a filler for paints and rubbers.

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