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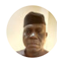

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Research Article
Quarry Waste as Secondary Raw Material for the Production of Surface Coating Agent-Paint

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Abstract
Background and Objective: Particularly, the processes of quarrying produce a lot of waste that has a lot of different mineral compositions, making it a potential secondary raw material for the formulation of surface coating products. The paradigm of a circular economy calls for a greater effort to locate suitable uses for this raw material. The goal of this investigation was to find low-cost local alternatives and adaptable raw materials for the manufacturing of water-based paint, with a focus on replacing the opacifier and filler without reducing the end product's quality. **Materials and Methods:** The quarry waste (QW) was pre-treated to produce treated quarry waste (TQW) and characterized for its mineral composition, surface morphology and crystallinity. Quixcoat, an emulsion-based paint, was prepared by NIS guidelines and subjected to physiochemical analysis. **Results:** The mineralogy analysis of the QW revealed the presence of anthophyllite, muscovite, quartz, garnet and albite, except orthoclase. The parameters of the Quixcoat were found to be within an acceptable range when compared to the Nigeria industrial standards and specifications. When compared to paint made from titanium dioxide and aluminum silicate, the properties of the Quixcoat, such as viscosity, density and hard drying time, showed improvements. However, it has a slightly high pH and a relatively low surface drying property, which are desirable properties for tropical paints. **Conclusion:** Hence, QW has the potential of serving as a potential secondary raw material for paint production.

Key words: Quarry waste, aggregates, fines processing, circular economy, mineral, paint

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The strong association between urbanization, economic growth and demand for mineral products has increased in the tonnage of waste generated over the last few decades, with no sign of a decrease[1]. Waste is a persistent issue in today's society and it must be addressed to ensure a sustainable future by SDG 12, which aims to reduce pollution and health consequences through ecologically sound waste management throughout the product life cycle and encourage waste prevention, reduction, recycling and reuse[2]. The waste from the quarry industry has been a major source of concern in most African nations, including Nigeria, due to poor management[3,4]. According to Afolabi *et al*.[5] the construction industry in Nigeria generates a significant amount of waste and up to 21-30% of a project cost overrun is attributed to materials waste[6] Globally, quarry waste (QW) accounts for approximately 20-25% of total waste generation, annually, equating to 238 million tones[7].

The QW typically has a particle size of less than 100 µm and a variety of mineral compositions[8]. According to Gbenu *et al*.[9] zircon, monazite, apatite, magnetite, ilmenite and riebeckite make up 61-65% of Nigeria's QW, whereas a smaller portion of QW is made up of rocks, soil and gravel. The shift from waste management to resource management is inevitable, this involves two strategies which are the use of renewable materials and the shift from a single-use linear economy to a circular economy. This has fueled the push for mineral recovery targets in several European Union (EU) member countries[10]. The specified mineral recovery objective has already been met in Austria and Germany, however, in Nigeria, there are weak statistics on waste generation and mineral recovery, as well as a lack of waste management policies and regulations[11-14]. Given the high mineral concentration of QW, it could be investigated for its potential use as an alternative raw material, which might cut costs without lowering the quality of the finished surface coating products such as paints[15].

Paints play a crucial role in architectural design and finishing, in addition to providing high-quality aesthetics for surface finishing, they also provide a protective coating that makes it possible for surfaces to withstand wear and tear from the elements of nature and the weather. Unfortunately, the paint industry in Nigeria has been plagued by several raw materials-related problems[16]. The majority of the paints contain volatile organic compounds, which help to improve the surface washability and durability and decrease the paint odour, but are toxic with short- and long-term adverse health effects[17,18]. In addition, Titanium Dioxide (TiO_2), a prime paint pigment and aluminium silica not only contribute to the high cost of paint but also hasten the rate at which painted surfaces disintegrate[19,20] and occupational and environmental health hazards[21].

According to Abdulsalam[22] global demand for paint and coatings rose by 3.7% annually to 54.7 million metric tonnes in 2020, with a value of $193 billion. The global market value of the paint and coatings industry decreased by roughly 6% to about 145 billion US dollars following the COVID-19 pandemic. Nonetheless, the industry's growth restarted in 2021, when it reached a market value of around 160 billion US dollars[23]. The sector's market value is expected to exceed 235 billion US dollars by 2029[24]. By 2025, Nigeria's paint industry is expected to reach $377 million (N135.80 billion) with a growth rate of 5% from its current estimated $268 million (N96.50 billion)[25]. In addition, the bank of industry (BoI) predicted that, in an ideal operating environment, local paint consumption would rise from 391.75 million liters in 2020 to an all-time high of 1,002.63 million liters by 2025[26]. This makes paint one of the largest consumers of natural resources.

As a result, alternative raw materials are required to address the cost, environmental and health concerns in the paint industry. Due to its enormous volume and unique mineral content, the waste produced by quarries is one of many potential alternatives that is particularly intriguing. Several scholars have emphasised the need of considering QW as secondary raw resources in a circular economy model. Following this concept, some authors reported the use of QW in pavement construction[27] partial sand replacement in construction material[28] and wood polymer composites[29]. Therefore, research into novel and the paint industry's need for better and cheaper raw materials and society's need for safe and economical waste disposal are being matched by innovative uses of QW. This investigation aims to find local alternatives and adaptable raw materials for the production of water-based paints that have the potential to cut costs without compromising quality, in addition to addressing waste management in the construction industry.

MATERIALS AND METHODS

Study duration: The study, which included the collection of the quarry waste, processing, paint formulation and analysis, was carried out from August, 2022 to February, 2023.

Study area: The quarry materials were collected from Kaduna State (10.3764°N, 7.7095°E) Nigeria and transported to the Industrial Chemistry Laboratory, Department of Chemical Sciences, Bingham University (8°57'14.6"N 7°41'55.0"E), Nigeria. The QW was stored at ambient temperature for further processing.

Reagents: Laboratory grade titanium dioxide, aluminium silicate, tetra potassium pyrophosphate, isobutyl vinyl ether, calcium carbonate, hydroxyethyl cellulose, ammonia, ethylene glycol, hydrogenated castor oil and yellow iron oxide were obtained from Merck chemicals, Germany.

Modification of locally sourced quarry site: The quarry waste was collected from manual quarrying site, packaged into 25 kg bags and transported to the laboratory. The collected quarry waste was oxidized with 36% peroxide solution, treated with sodium hypochlorite and washed with deionized water. Then, oven dried for 72 hrs at 100°C and mechanically de-sizing. Then, subjected to flame pyrolysis at a temperature of 3000°C electric arc and sieved to defined mesh sizes to achieve the optimal non-porous particle size, surface area and a density.

Characterisation of modification locally sourced quarry waste: The morphologies and surface structures of modified material were characterized with Hitachi S-4800 Scanning Electron Microscope (SEM). The functional groups were determined on PerkinElmer Spectrum 400 FT-IR (Waltham, MA, USA) scanned between 4000 and 400 cm^{-1}. While an X-ray Fluorescence (XRF) spectrometer (MESA-50 EDXRF Analyzer, Horiba Instruments, New Jersey, USA) was used to identify the composition.

Paint formulation: A standard procedure for the formulation of paint was used to prepare the paint with the label "Quixcoat", however, the treated quarry waste (TQW) was used in place of the conventional opacifier (titanium dioxide) and filler (aluminum silicate) in the standard paint formula. A high-shear impeller mixer with a medium speed was used to add 1.20 g of tetra potassium pyrophosphate to about 40 g of water. After that, 0.60 g of isobutyl vinyl ether and 60 g of TQW were added. To complete the first stage of dispersion, 360 g of calcium carbonate and 40 g of water were added and the mixture was stirred for 60 min. After continuous blending for 60 min, 5.4 g of hydroxyethyl cellulose, 24 g of water and the remaining 0.6 g of isobutyl vinyl ether were added and the mixture was mixed for another 25 min at a setting of 5 rpm to complete the second scattering phase. After that, 0.6 g of ammonia, 6.0 g of ethylene glycol, 1.2 g of hydrogenated castor oil, 200 g of TQW, 0.05 g of yellow iron oxide and the remaining 24 g of water were added and the mixture was stirred once more for 30 min at a fixed speed of 5 rpm.

Paint physicochemical properties: The American Society for Testing and Materials' international standards were used to characterize the physicochemical properties of the emulsion paint (Quixcoat). However, the soluble lead content, resistance to fungal growth, coarse particles and foreign matter were all carried out per standards[30].

Gloss at 85°C head: At 85°C following the ASTM D523 standard test method for specular gloss, a gloss meter (Tri Gloss Model: G68, Bangalore, India) was used to directly measure the gloss of the paint, which is refer to the paint's sheen level. The paint will be more reflective the higher the sheen level.

Determination of viscosity: Using the Standard Test Method for Viscosity proposed by ASTM D 1200-10, a viscosimeter (SVM 3000 Stabinger, Ashland, VA) was used to obtain the viscosity profile of the Quixcoat.

Determination of pH: According to ASTM-D 1208-90 standard test methods for common properties of certain pigments, a benchtop meter pH meter (Seven Excellence S400-Basic, METTLER TOLEDO) was used to determine the degree of acidity or alkalinity of the Quixcoat. About 35 mL of Quixcoat was carefully blended with an equal volume of distilled water in a beaker. The probe was inserted into the solution and a reading was taken.

Drying time: The drying time was done following the ASTM-D1640 Standard Test Methods for Drying, Curing, or Film Formation of Organic Coatings. Quixcoat was applied to a clean, dry wall and monitored and the time it took for the film to surface and hard-dry was recorded in minutes.

Durability: The paints produced were subjected to a durability test to ascertain how long they can stand before deteriorating. The paint produced by substituting the opacifier and filler with TQW was allowed to stand for 21 days after which they were observed and a quality test to determine viscosity, density, pH and drying time was performed.

Statistical analysis: The Analysis of Variance (ANOVA) GraphPad Prism software version 5.1 was used to analyze the data, which were expressed as Mean±Standard Error of the Mean (SEM). A significance level of $p<0.05$ was deemed statistically significant.

Spreading rate: The percentage volume solids value and dry film thickness (DFT) values of the Quixcoat, were utilized in the calculation of the spreading rate (Eq. 1) adapted from the Dulux protective coatings method[31]:

$$\text{Spread rate (m}^2\text{/L)} = \frac{\text{Volume solids (\%)}}{\text{Dry film thickness}} \times 100$$

Opacity: According to ASTM D6762-18, the Standard Test Method for determining the hiding power of paint was determined. The paint was applied on the sheen opacity panel and the rating was done accordingly.

Adhesion to substrate: It is one of the most important properties of the coating. The difficulty of the coating to detach from the substrate is referred to as adhesion. In line with ASTM D 3359 Test Method A, an X-cut to the substrate is made through the film using a carbide tip tool. The pressure-sensitive tape was used to cover the cut. The tape was smoothed into place over the incisions using a pencil eraser. The tape was quickly removed by bringing it back over itself at a 180° angle. Adhesion was graded on a scale of 0 to 5.

RESULTS AND DISCUSSION

Organic component analysis: The quarry waste, treated quarry waste, Quixcoat and titanium dioxide-aluminium silicate-based paint functional group spectra were depicted in Fig. S1(a-d). At 1133 cm^{-1} and 760 cm^{-1}, the QW and TQW displayed similar bands with varying peak intensities Fig. S1(a-b). The absence of a high-polarity bond is the cause of the decrease in peak intensity observed in TQW. The obtained spectra demonstrated the absence of organic moieties in QW and TQW, Consequently, QW and TQW are ineffective as microbial growth substrates. When used in the formulation of a surface coating agent, this will increase the storage potential and stability.

In the spectra of titanium dioxide-based paint and Quixcoat, two key vibrational bands were observed around 3332 cm^{-1} and 1640 cm^{-1}. The FTIR spectra exhibit a medium acetate ester absorbance band at 1640 cm^{-1} due to the carbonyl group of polyvinyl acetate and hydroxyl group of ethylene glycol used in the formulation process. The Fig. S1(c and d) did not indicate any distinction in bonding order or functional group type, this demonstrated that the utilization of TQW in paint formulation proceeds via a physical process[32]. The orientation of the spectra suggested that there is no chemical interaction between the TQW and the conventional paint raw material in the formulation of Quixcoat. As a result, new compounds with potential negative effects on human health and the environment were absent.

Metal analysis: The findings of the metal analysis were presented in Table 1, which showed the seven primary elements with percentage concentrations below $\geq$1.10.

The QW revealed a significant concentration of cerium (10.97%) and rhodium (20.01%). However, when the QW was subjected to a series of bleaching and oxidation, it yielded a large quantity of cerium (IV) oxide (13.33%), rhodium sesquioxide (23.00%), aluminum oxide (15.01%) and silica (8.82%) as compared to the untreated QW. This might be attributed to organic and oxidizable substances leached during the treatment of the QW. Cerium (IV) oxide has been found in studies to improve pigment photostability by providing lightfastness and preventing transparent polymers from darkening in the sunshine[33] whereas, rhodium sesquioxide is known to protect surfaces from environmental damage[34]. Additionally, the high concentrations of aluminium oxide and silica point to the TQW's potential use as fillers. This was consistent with the research conducted by of Sadej and Andrzejewska[35] regarding the use of a silica/aluminum oxide hybrid filler in photocurable composites. Tungsten, zirconium and manganese were among the other main minerals whose percentage compositions did not change considerably after the treatment, because the bulk of these minerals were present in their unoxidisable state.

Mineral characterization: The mineral composition of treated quarry waste, untreated quarry waste, Quixcoat and titanium dioxide-aluminium silicate-based paint prepared to Nigerian industrial standards was examined using X-ray Diffraction Analysis (XRD). The diffractograms were presented in Fig. 1(a-d).

In general, the QW and TQW diffractograms indicated a large quantity of crystallite material in the samples, but the paints revealed a roughly comparable composition with slight amorphous characteristics. The analysis results of QW and TQW was shown in Fig. 2(a-b), demonstrated the presence of Anthophyllite (Mg_2 Mg_5 Si_8 O_{22} $(OH)_2$), Muscovite (KAl_2 $(FeOH)_2$), Quartz (SiO_4), Garnet ($[Mg,Fe,Mn]_3Al_2(SiO_4)_3$) and Albite ($NaAlSi_3$ O_8) except orthoclase a tectosilicate mineral ($KAlSi_3$ O_8) which forms igneous rock. The absence of orthoclase in TQW is comprehensible given the fragility of tectosilicate minerals at high-temperature as reported by Ni *et al.*[36]. The XRD result obtained in the study by Amin *et al.*[37] involving the utilization of Gebel Attaqa Quarry Waste in the production of Single Fast Firing Ceramic Wall Tile is in good agreement with the mineral analysis of the QW.

Calcite ($CaCO_3$), quartz and muscovite were found in both the titanium dioxide-aluminium silicate-based paint and Quixcoat's mineral content was shown in Fig. 2(c-d). Additionally, the titanium dioxide-aluminium silicate-based

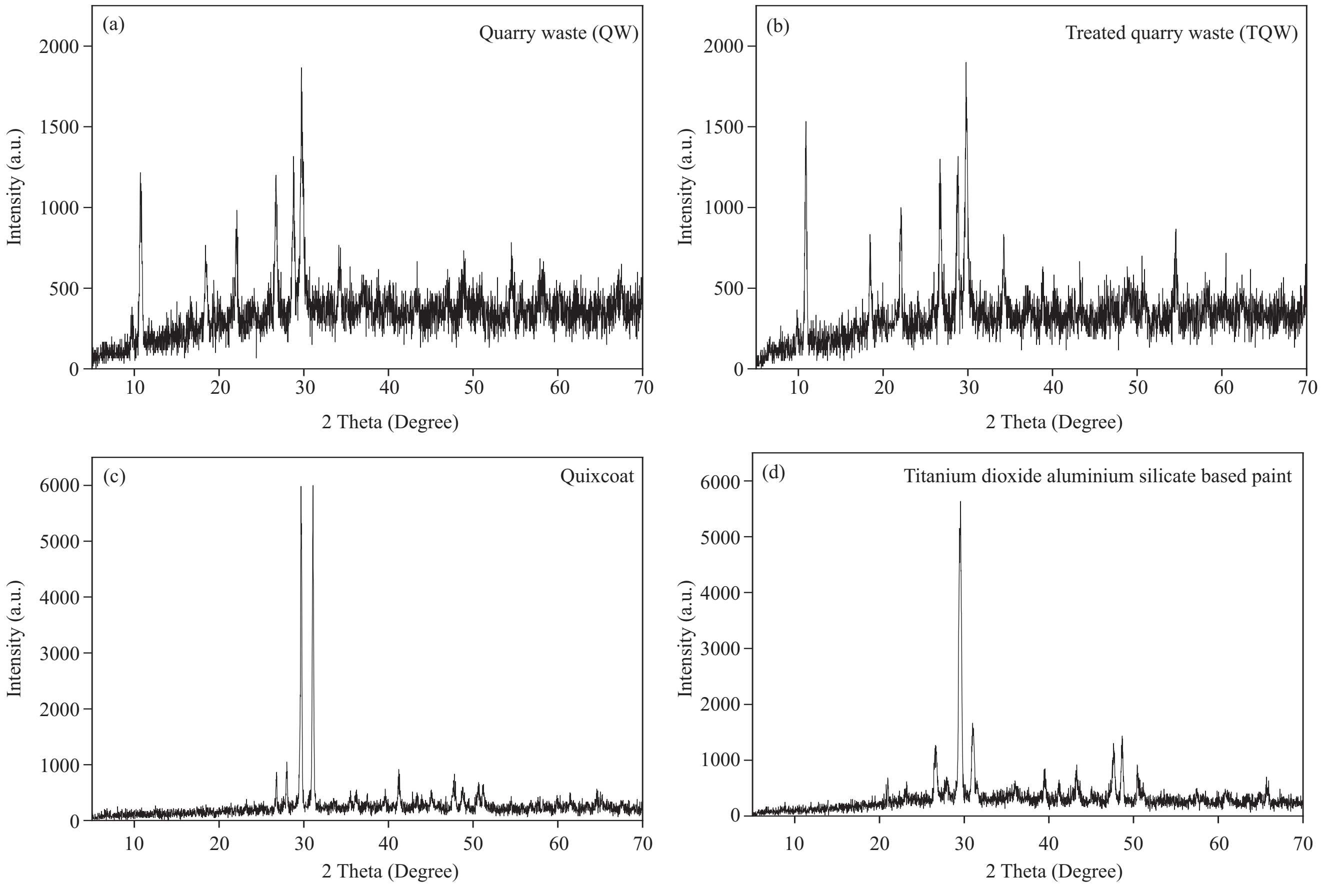

Fig. 1(a-d): Spectra of (a) Quarry waste (QW), (b) Treated quarry waste (TQW), (c) Quixcoat and (d) Titanium dioxide-aluminium silicate-based paint is submitted alongside this manuscript

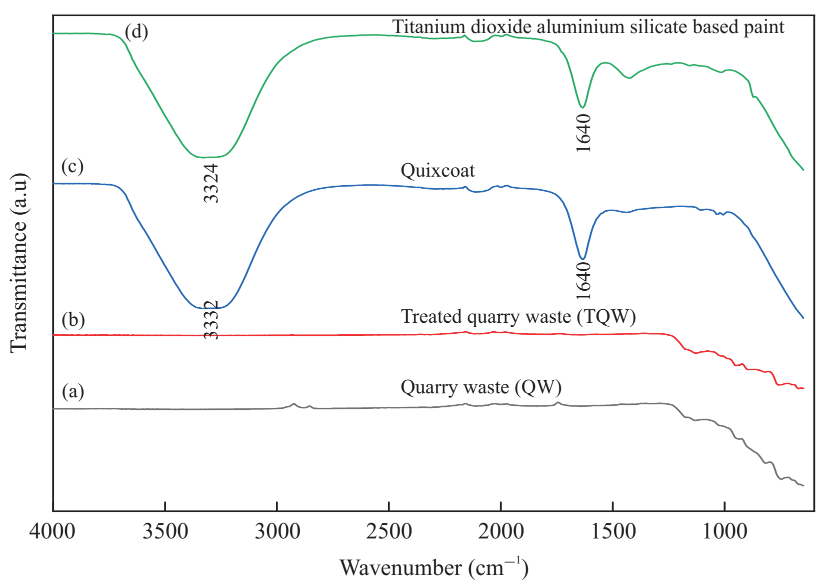

Fig. S1(a-d): Diffractograms of (a) Quarry waste (QW), (b) Treated quarry waste (TQW), (c) Quixcoat and (d) Titanium dioxide-aluminum silicate-based paint

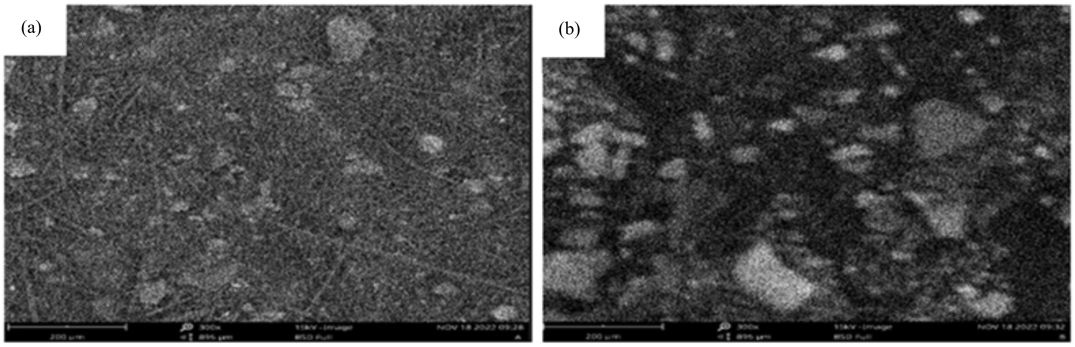

Fig. 2(a-b): Micrographs of the (a) Quarry waste and (b) Treated quarry waste

Table 1: Major components in the untreated quarry waste and treated quarry waste (%)

Samples	CeO_2 (%)	TiO_2 (%)	ZrO_2 (%)	Al_2O_3 (%)	SiO_2 (%)	Rh_2O_3 (%)	MnO (%)
QW	10.97	1.04	5.43	08.11	4.34	20.01	1.10
TQW	13.33	2.00	5.68	15.01	8.82	23.00	1.34

QW: Untreated quarry waste and TQW: Treated quarry waste

Table 2: Paint properties

Parameter	Quixcoat	Standard paint parameters
Soluble lead content (ppm)	7.8 ppm	≤90
Resistance to fungal growth(%)	1.3%	≤3%
Specular glass @85° (sheen) flat finish	3.9	≤15
Drying properties (min)		
surface dry	41 min	≤20
Hard dry	66 min	≤120
Spreading rate (m^2/L /min)	9.1 m^2/L/min	8
pH value	9.87	7.00-9.00
Viscosity poises (min) @27°C+2°C	51.5 min	6.0 min
Coarse particles and foreign matter (% w/w)	0.35	≤1
Opacity(%)	0.98%	0.95-1%
Adhesion strength (ASTM classification)	5B	0B-5B

paint contained dolomite $(CaMg(CO_3)_2$ and orthoclase. The occurrence of dolomite and orthoclase is due to the inclusion of industrially processed material in the preparation of the NIS paint. According to Gettens *et al.*[38] this was in line with the mineralogy analysis of standard paint as reported in the study on understanding calcium carbonate and dolomite as extenders.

Surface morphological analysis: The micrographs of the quarry waste and treated quarry waste analyzed at 200 µm at a magnification ×300 and the working distance of 859 µm was shown in Fig. 2. The micrograph of quarry waste displayed a microstructure and a general glassy matrix in which different crystal phases are present[39]. The treated quarry waste, on the other hand, is lumpy and has a large, uneven shape. The differences in surface morphology are due to the presence of extraneous materials such as organic material and fine silicate in the QW which were removed by the bleaching and oxidization steps.

Architectural implication of quixcoat: The results for the paint properties were shown in Table 2. Quixcoat met the recommended standards for its soluble lead content, resistance to fungal growth, specular glass, spreading rate, pH, coarse particles, opacity, substrate adhesion and foreign matter. When applied to surfaces, Quixcoat has the ability to reflect light and heat radiation, keeping homes cool, as evidenced by the 3.9@85° (sheen) specular glass flat finish. Further, stains can also be easily scrubbed off, providing surfaces that encourage high water runoff. This specular glass flat finish value may be linked to the crystallinity and quartz contents. The Quixcoat's hard drying property (66 min) and viscosity (51.5 min) are significantly improved compared to the Nigerian industrial standards recommended 120 and 6 min, respectively. However, Quixcoat surface drying time was reported to be 41 min as opposed to 20 min as a result of the high viscosity value reported. According to Talbert[39], the paint dries more quickly lower its viscosity.

The activities of microbes, which are unable to feed on the cellulose in aerosol due to their synthetic nature, are responsible for the resistance to fungal growth. They also stop the growth of mold and algae, which deface surfaces and reduce the capability of building materials. Quixcoat is resistant to fungi growth and are most suitable for use in regions with high temperatures and humidity conditions such as Nigeria. As a result, production costs will be reduced because there won't be a need for a preservative agent in the paint.

The modification increased the basicity of the paint as the paints produced with the modified QW had a pH value of 9.87 compared with 7.00-9.00 for paint produced as recommended by Nigerian industrial standards. Studies by Schubert *et al.*[40] and Tang *et al.*[41] have shown how the presence of metal ions may modify the pH of a system, consequently, the concentration of hydroxide ions when mineral-rich TQW is used as an alternative material in paint formulation. Table 2 showed the results of an evaluation of Quixcoat adherence to the surface of a substrate. Adhesion was assessed using the ASTM D3359-17 standard, with a scale of 0B-5B representing the degree of adhesion from the worst to the best. A score of 5B was obtained, which may indicate that the coatings (Quixcoat) have a lower surface energy than the substrate, allowing for good wetting and adhesion[42].

Benefits and limitations: The QW is a cost-saving raw material that easily substitutes the expensive titanium dioxide and aluminium silicate used in convectional paint formulas. The main limitation of this substitute is the inconsistency of quarry waste, which is location and mineral dependent. This will alter the consistency of the finished paint.

Further implementations: The outcome of this study implies that there will be an increase in the demand for QW as a secondary raw material in the production of paint, as it easily replaces the traditional opacifier (titanium dioxide) and filler (aluminum silicate) in the standard paint formula. This will reduce the cost of production, however, this also implies the attendant increase in the environmental effects such as noise pollution and contamination of water sources due to industrial tailing.

Future recommendations of study: Quarry waste is in high seasonal demand. It's primarily used in the construction industry as a chipping component of asphalt, stone-base material and stone dust. The Nigerian Miners' Cooperation needs to develop regulations that will protect the environment, as this study has revealed that QW will not be required on a seasonal basis but on a constant and regular basis.

CONCLUSION

The major finding of this study revealed that QW can be modified to meet the local paint consumption need as well as address the cost, environmental and health concerns in the paint industry. A key driver for a shift from waste management to resource management is based on the paradigm of a circular economy, thereby creating jobs and reducing the amount of capital allocated to QW management. On the other hand, the inability of microbes to thrive on the Quixcoat painted surface and the specular glass flat finish value make Quixcoat the surface coating agent of choice in high humidity and temperature region like Nigeria. In terms of cost, the substitution of the traditional opacifier and filler with mineral-rich-TQW has the potential of addressing the high production cost concern of local paint producers. From an architectural perspective, Quixcoat has an outstanding re-coating capability, including no color separation, sagging, flaking, or cracking and a high opacity when stored for an extended period, as specified by NIS 278:1990.

SIGNIFICANCE STATEMENT

The significant of this study was to discover low-cost local options and flexible raw materials for the production of paint, with an emphasis on substituting the opacifier and filler without lowering the standard of the finished surface coating product. The mineralogy of the quarry waste revealed the presence of a lot of orthorhombic and plagioclase minerals. These minerals are used as insulation and building materials and they are sometimes added to paints and ceramic glazes to give them shine. The paint's viscosity, density and hard curing time were found to be within acceptable ranges when compared to industrial specifications.

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