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Quality Improvement of Off-Spec Coarse Aggregates with the Use of Unsaturated Polyester Resin

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ABSTRACT

This study aims to modify off-spec limestones using resin to enhance their mechanical properties. The modified limestones are intended for road construction. The mechanical properties are accessed through Aggregate Crushing Value (ACV), Aggregate Impact Value (AIV), Los Angeles (LA) abrasion, water absorption, and microstructure analyses. Then, the modified limestones are exposed to deterioration to resemble real-life altering conditions on highways. The exposure conditions consist of the direct immersion of water, 3% NaCl solution, and 1% HCl solution, and thermal stress cycles. This was followed by quality test analyses for comparisons and inferences. The samples were immersed in water, NaCl solution, and HCl solution for 63 days, with each cycle consisting of 3 days of immersion and four days of air drying. Nineteen thermal stress cycles were carried out, with 18 hours of immersion in NaCl solution and 6 hours of heating in an oven at 60 degrees. The findings show significant improvement in crushing value, impact value, and abrasion resistance of aggregates after surface treatment with resin. Microstructure analyses using SEM revealed that the treated limestone had a rougher surface texture, indicating enhanced bonding in concrete, facilitating chemical reactions, improving mechanical interlocking, and ultimately enhancing the overall performance and durability of the concrete structure.

1. INTRODUCTION

This concrete production increases with development. In general, approximately 60–75% of concrete volume is comprised of aggregates [1]. Hence, the global consumption of construction aggregates has increased [2, 3].

According to the United Nations Department of Economic and Social Affairs, urban areas will accommodate 68% of the world's population by 2050 [4]. It is forecasted that the global construction aggregate will reach 62.9 billion metric tons by the end of 2024, which will be up from 43.3 billion metric tons in 2016 [4]. To sustain the country's economic development, the mining and quarry industry must ensure a sufficient and consistent provision of raw materials to the construction industry [5-7]. The consequence of insufficient building supplies will lead to higher construction costs, which will then be passed on to end users, jeopardising national development [8].

Natural aggregates in Malaysia are widely used in the concrete production industry. However, in some situations, construction aggregates do not meet project or industry specifications regarding one or more properties such as soundness, water absorption, abrasion resistance, or contamination with chlorides and sulphates. Adhered mortar on aggregate is the main factor affecting the mechanical properties of recycled concrete aggregates (RCAs), such as density, porosity, and water absorption [9-13].

In some other situations, the available aggregates are reactive aggregates that, when used without prevention measures, will deteriorate concrete members. According to JKR 2020, crushed granite shall only be utilised for work below ground level. The source of the problems with concrete below ground level is studied, and one of the issues is a chemical attack on limestones below the ground level.

To comply with the project specification, the proper type of aggregates needs to be imported from another location, which would increase cost and carbon footprint [14]. Thus, it is crucial to promote sustainability and lower carbon footprint by resolving one or more off-spec aggregate properties using suitable treatments.

Existing methods for enhancing aggregate performance, such as mechanical processing, chemical coatings, thermal treatment, and polymer/bitumen coatings, exhibit several limitations, underscoring the need for an alternative approach. Mechanical processing improves grading but does not address fundamental issues such as high porosity and low durability

[13]. Chemical coatings enhance durability but have limited impact on mechanical strength and require precise application [10]. Thermal treatments, while effective in removing adhered mortar, are energy-intensive and can induce micro-cracking, compromising long-term performance [3]. Polymer and bitumen coatings improve moisture resistance but may degrade under UV exposure and extreme temperatures, limiting their effectiveness [14].

Untreated off-spec aggregates often fail to meet industry specifications due to high water absorption and poor mechanical properties, leading to premature pavement failure [15, 16]. The proposed unsaturated polyester resin treatment addresses these limitations by enhancing mechanical strength (ACV, AIV, LA abrasion), reducing water absorption, and improving resistance to environmental degradation while requiring minimal energy input. This study builds on existing research by demonstrating the feasibility of resin treatment as a sustainable and cost-effective solution for road construction applications.

Maximising the utilisation of natural off-spec construction aggregates while ensuring their optimal performance guarantees a reliable supply of raw materials to the construction sector [17].

From a global perspective, similar issues arise, particularly in regions with high construction activity. The demand for natural concrete aggregates often outpaces availability, increasing costs and environmental impacts [6]. Moreover, concerns over the carbon footprint and ecological consequences of aggregate extraction are rising globally. The significance of sustainability in concrete construction stems from the substantial environmental impact of the construction industry, mainly attributed to the extensive utilisation of concrete as a fundamental construction material [18]. Implementing sustainable practices, such as utilising off-spec aggregates, can help address these challenges and promote a more environmentally friendly construction industry worldwide. Recycling concrete is necessary because it helps to promote sustainable development by reducing the disposal of demolition [19]. Using recycled aggregates sourced from CDW in concrete making provides a way to recover the CDW as a raw building material and minimise the natural mineral resources [20]. Achieving widespread acceptance and implementation of alternative aggregates requires concerted efforts in research and development, industry collaboration, and policy support.

More research is needed on the quality improvement of off-spec coarse aggregates through thermosetting surface treatment using unsaturated polyester resin. Due to their excellent cohesion with other materials, polyester and vinyl ester resins are more accessible to produce and are more typically used in vital industries [21]. Resins are selected for surface treatment as they are non-corrosive, increase concrete durability, and lower maintenance and repair expenses.

The objectives of this research are:

•To improve the mechanical performance (crushing strength, impact resistance, abrasion resistance) and durability of off-spec limestone aggregates through surface treatment with unsaturated polyester resin.

•To evaluate the effects of resin treatment on water absorption and microstructure changes in the modified aggregates.

•To assess the long-term durability of treated aggregates under simulated environmental conditions, including exposure to NaCl, HCl, and thermal stress cycles.

•To compare the performance of resin-treated aggregates with standard granite and untreated limestone aggregates based on industry specifications for road construction.

This study investigates the utilisation of natural concrete aggregates in the local construction industry. The intrinsic parameters of off-spec aggregates, such as density, water absorption, ACV value, AIV value, and micro-texture, are evaluated under various exposure conditions. The treatment technique's effectiveness is assessed by analysing the aggregates' SEM imagery. The potential advantages in concrete production are analysed by exploring the incorporation of off-spec aggregates as alternatives to standard natural aggregates. In short, the research promotes the optimisation of off-spec aggregate usage in construction projects, thereby ushering the concrete industry towards enhanced sustainability and efficiency.

2. METHOD

This study comprehensively evaluates resin-treated aggregates, assessing their structural integrity, durability, and potential application in pavement engineering. The findings will help determine whether unsaturated polyester resin modification is a viable alternative for improving the performance of off-spec aggregates in sustainable road construction.

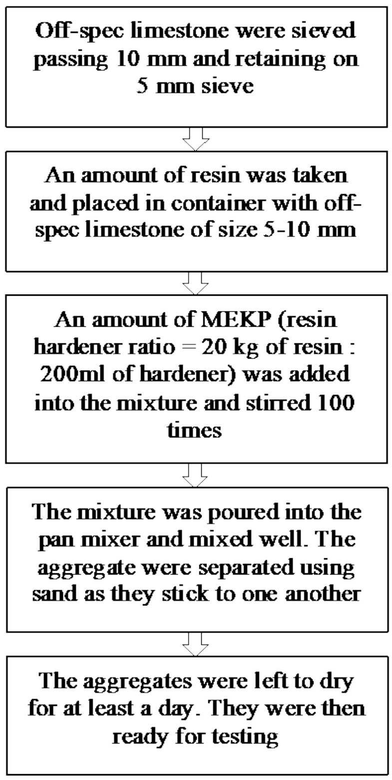


Figure 1. Flowchart of sample preparation

As shown in Figure 1, the treatment of off-spec limestone aggregates in this study involved the application of unsaturated polyester resin combined with a hardener to facilitate curing and fine sand to prevent particle adhesion during the coating process.

To evaluate the durability and performance of the resin-treated aggregates, two categories of samples were prepared:

•Non-exposed samples consist of untreated limestone, resin-treated limestone, and standard granite (used as a control for comparison).

•Exposed samples – Comprising resin-treated limestone subjected to various environmental stressors to assess durability under real-world conditions.

The exposure conditions included:

•Chemical immersion tests: Samples were submerged in water, 3% sodium chloride (NaCl), and 1% hydrochloric acid (HCl) solutions for 63 days to assess resistance to moisture, chemical degradation, and salt-induced damage. Each cycle lasted three days of immersion and four days of air drying under laboratory conditions.

•Thermal stress cycles: To evaluate resistance to temperature fluctuations, treated limestone underwent 12 thermal cycles, each consisting of 18 hours of immersion in 3% NaCl solution at room temperature, followed by 6 hours of drying at 60°C in an oven.

Figure 2 is the sample used in this investigation before treatment. To assess the mechanical performance and durability of both exposed and non-exposed samples, the following laboratory analyses were conducted:

•Aggregate Crushing Value (ACV): Determines the compressive strength and resistance to crushing under applied loads. Lower ACV values indicate stronger and more durable aggregates, essential for high-traffic road applications [15].

•Aggregate Impact Value (AIV): Evaluates the resistance of aggregates to sudden impact or shock loading. A lower AIV signifies higher toughness and durability, which is crucial for withstanding vehicular loads [22].

•Los Angeles (LA) Abrasion Test: Measures the abrasion resistance of aggregates due to mechanical wear. This test is particularly important for road surface materials, as higher abrasion resistance ensures longer pavement lifespan and reduced maintenance costs [23].

•Water Absorption Test: Determines aggregates' porosity and moisture susceptibility, which directly influences their durability and long-term performance in construction applications [6, 13].

•Scanning Electron Microscopy (SEM) Analysis: Examines the microstructural changes in treated aggregates, particularly surface roughness and bonding characteristics. A rougher surface texture typically enhances mechanical interlocking with cement paste, leading to improved strength and in concrete applications [24].

Figure 2. The samples before treatment with resin

3. RESULT

3.1 Industry specifications

As the modified limestones are for the use of road construction, the industry specifications by JKR are presented in Table 1. These specifications verify if the sample aggregates are suitable for specific concrete uses in road construction.

Table 1. Industry requirements for coarse aggregates to be used as wearing surfaces and other concrete uses [14]

Industry Specifications for Coarse Aggregates	
Tests	**Criteria**
ACV	At most, it should be 45% for aggregates used in concrete, not for wearing surfaces. Should not exceed 30% for wearing surfaces.
AIV	It should not exceed 45% for aggregates used in concrete other than for wearing surfaces. Should not exceed 30% for wearing surfaces.
LA abrasion value	Concrete should not exceed 30% in weight when used on wearing surfaces. It should not exceed 50% in weight for other concrete uses.
Water absorption	It should be at most 5%.

The strength of coarse aggregates is determined by the Aggregate Crushing Value test. A lower ACV indicates that the aggregate sample has a stronger resistance to breaking or deforming under compressive stresses [22, 23]. Essentially, it indicates that the aggregate has greater strength. The ACV values for all samples in this study are tabulated in Table 2.

Table 2. ACV results for all samples

Samples	Exposure Condition	ACV Results (%)
Granite	Nil	45
Limestones	Nil	41
Modified limestones	Nil	17
	Water	44.3
	3% NaCl	41.8
	1% HCl	42
	Heat cycles	42

The ACV for modified limestone without exposure was 17%, a significant improvement from 41% in untreated off-spec limestone. This reduction indicates enhanced mechanical strength and durability, making the treated aggregates more resistant to compressive loads. Compared to standard granite aggregates, the modified limestone exhibited superior crushing resistance, reinforcing its suitability for high-durability pavement applications.

Similar improvements in ACV through surface modification techniques have been reported in previous studies. Aggregates with ACV below 30% are ideal for wearing surfaces, aligning with industry standards [14]. The findings confirm that unsaturated polyester resin treatment effectively strengthens off-spec aggregates, making them a viable alternative for road construction [22, 23].

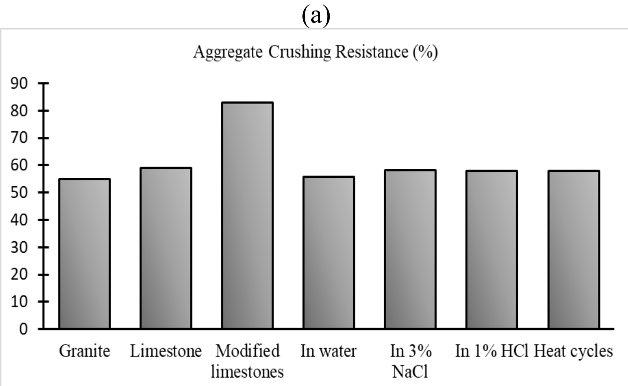
However, the ACV of modified limestones increased after exposure to various environmental conditions, with the highest increment observed in samples immersed in water. The ACV of samples exposed to 1% HCl solution and thermal cycles reached 42%, while those subjected to 3% NaCl exposure recorded 41.8%, representing only a 0.5% increase from the untreated off-spec limestone (41%).

Despite this increase, all ACV values remained below 45%, which, according to BS 812-110 (1990) and JKR (2020) standards, indicates suitability for general concrete applications but not for wearing surfaces [14]. Table 1 shows aggregates intended for wearing surfaces should have an ACV below 30%. The modified limestone initially met this requirement (ACV = 17%), making it viable for high-durability road layers. However, prolonged exposure to NaCl, HCl, and heat cycles significantly increased aggregate fragmentation, rendering the material unsuitable for wearing surfaces.

These findings align with studies that reported similar aggregate strength degradation under chemical and thermal exposure [22, 24]. Additionally, polymer-coated aggregates can initially improve durability but may deteriorate over time when subjected to prolonged environmental stressors [15].

Following surface treatment, the ACV of off-spec limestone decreased by 62.2%. This reduction is attributed to fewer aggregates passing through the 2.36 mm sieve after the applied load. The treated limestone exhibited more excellent crushing resistance compared to untreated limestone and granite, primarily due to the binding and adhesive properties of the resin.

(a)



(b)

Figure 3. (a) The physical observation after the ACV test; (b) The aggregate crushing resistance of the samples

Following the ACV test, the aggregates remained bound together, as depicted in Figure 3(a). The aggregate crushing resistance, calculated as (100 – ACV): is presented in Figure 3(b). The results indicate that modified off-spec limestone exhibited the highest crushing resistance, reaching 83%, demonstrating a significant enhancement in mechanical strength. This indicates that only 17% of the material was crushed under the applied load, reflecting improved aggregate integrity and reduced susceptibility to fragmentation. This is crucial for ensuring long-term durability in pavement applications [25].

3.2 AIV

Table 3 records all the test samples' AIV. Aggregates with lower AIV values indicate superior quality and are less likely to break or fracture when subjected to impact [15].

Table 3. AIV results for all samples

Samples	AIV Results (%)
Granite	14
Limestones	11
Modified limestones	5
In water	7
In 3% NaCl	8
In 1% HCl	7
Heat cycles	10

The AIV of modified limestone under unexposed conditions was recorded at 5%, demonstrating a significant improvement in impact resistance compared to raw off-spec limestone with an AIV of 11%. This suggests that resin treatment effectively enhances aggregate toughness, making it more resistant to impact forces than untreated limestone and standard granite. However, exposure to extreme environmental conditions increased AIV, with heat cycles causing the highest increment at 10%. Despite this increase, the AIV of modified limestone remained lower than that of untreated limestone and standard granite, indicating that the resin-treated aggregates retained superior impact resistance. This finding aligns with previous research highlighting the role of polymer-based surface treatments in improving aggregate durability [26].

3.3 LA abrasion

The aggregates used in the surface course of the highway pavements are subjected to wearing due to traffic movement.

As highway pavement surface coarse aggregates undergo wear and tear from vehicular movement, the interaction between the pneumatic tyres and the road surface instigates an abrasion of the road aggregates, primarily due to soil particles [27].

The LA abrasion test results in this study are tabulated in Table 4. The abrasion value was reduced in modified limestones (17%) as compared to standard granite (22%) and raw off-spec limestones (21%). This indicates improvement in mechanical quality after surface treatment.

Table 4. Aggregate abrasion values for all samples

Samples	LA Abrasion (%)
Granite	22
Limestones	21
Modified limestones	17
In water	10
In 3% NaCl	8
In 1% HCl	6.2
Heat cycles	7.2

The results further presented improvements for the treated off-spec limestones under all the exposure conditions. This was unexpected and requires further study for verification. The abrasion value is required to be less than 30% by weight for concrete to be used for wearing surfaces and 50% by weight for other concrete uses. All the samples recorded abrasion values lower than 30%, with the highest abrasion value at 22% in standard granite. Thus, the aggregates are suitable for all concrete uses regarding LA abrasion.

3.4 Water absorption

The water absorption values for all the samples are presented in Table 5.

Table 5. Water absorption (%) of all the samples

Samples	Water Absorption (%)
Granite	29.45
Limestones	18.49
Modified limestones	17.19
In water	2.5
In 3% NaCl	2.95
In 1% HCl	0.69
Heat cycles	1.15

Based on the industry requirement, coarse aggregates should not absorb water by more than 5%. However, the standard granite and untreated off-spec limestones have more than 5% water absorption. It is observed that the water absorption can be lowered in off-spec limestones after treatment with unsaturated polyester resin. Though the surface treatment using the thermosetting method lowered the absorption value, it did not meet the industry requirement. Further, when exposed to different conditions, the treated limestones recorded a higher reduction in water absorption than the samples that were not exposed to any conditions. The samples exposed to different conditions meet the industry requirement with a water absorption value lower than 5%. For instance, when exposed to 1% HCl solution, treated limestone recorded the lowest water absorption at 0.69%. Though it is expected that the mechanical properties of treated limestones will be reduced with exposure to extreme conditions (prolonged exposure to NaCl, HCl, and heat), the findings in terms of water absorption are unexpected.

4. SEM IMAGERY

The morphology of the aggregate particles can substantially impact the workability, strength, and durability of the concrete. SEM imagery was done on all the samples.

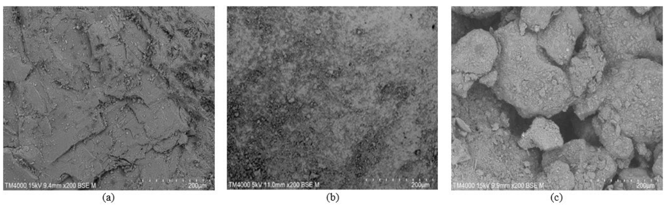


Figure 4. (a) The micrograph of standard aggregate (granite) at resolution 200; (b) The micrograph of limestone at resolution 200; (c) The micrograph of treated limestone at resolution 200

The micrographs for granite, limestones and treated limestones are illustrated in Figures 4(a)-4(c), respectively. Both granite and untreated limestone displayed a smoother surface than treated off-spec limestone. They consisted of a smaller and more uniform shape of grains.

The micrograph of treated off-spec limestone shows particles of varying sizes with a rougher surface, likely due to sand adhesion during treatment. A rougher surface enhances the bond between fine aggregates and asphalt, improving pavement performance [28, 29]. Aggregate surface texture significantly affects concrete properties. Rougher surfaces strengthen the bond with cement paste, increasing mechanical performance, while smoother surfaces enhance workability [30]. Incorporating rough-textured aggregates can optimise mix durability [31].

Figures 5-8 compare treated off-spec limestone with samples exposed to different conditions, highlighting surface modifications and degradation mechanisms.

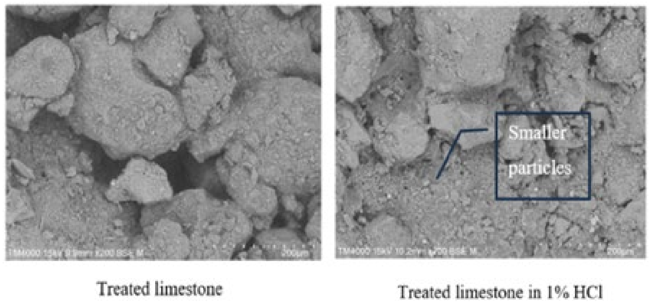


Figure 5. The comparison between micrographs of treated limestone with the treated limestones under 1% HCl exposure conditions

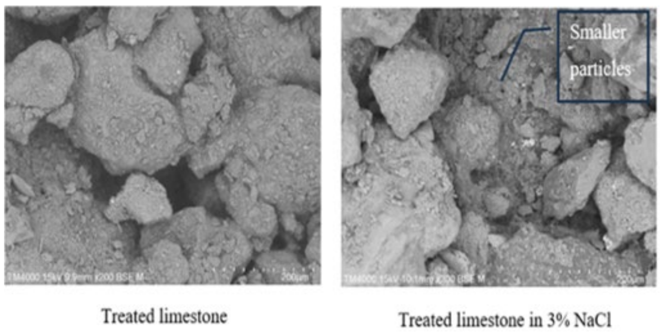


Figure 6. The comparison between micrographs of treated limestone with the treated limestones under 3% NaCl exposure conditions

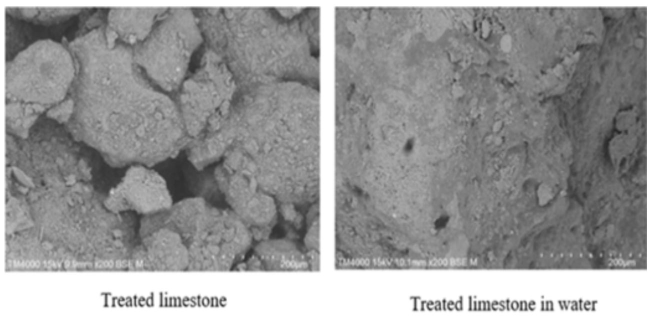


Figure 7. The comparison between micrographs of treated limestone with the treated limestone underwater exposure condition

Figures 5-8 show that these results align with studies on environmental impacts on aggregate durability. However, the long-term performance of resin-treated aggregates under real traffic and weather conditions remains uncertain. Future research should assess their behaviour in asphalt mixtures, prolonged exposure to mechanical loads, and environmental stresses. Additionally, evaluating compatibility with various asphalt binders would support their practical application in pavement engineering.

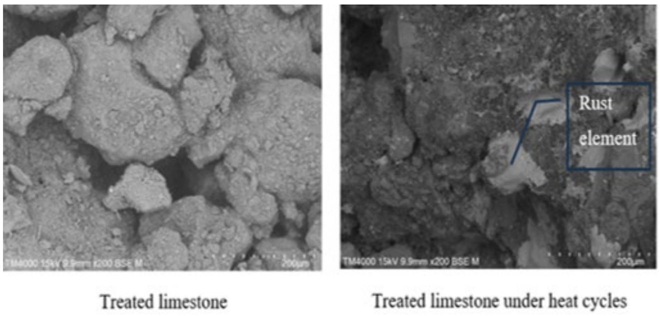


Figure 8. The comparison between micrographs of treated limestone with the treated limestones under heat cycle conditions

The micrograph results indicate a reduction in distinct-sized particles on treated limestone under various exposure conditions compared to treated limestone with no exposure. This effect is particularly noticeable in limestone subjected to water immersion and heat cycles (Figures 5-8). Additionally, smaller particles are increased in limestone exposed to 1% HCl, 3% NaCl, and heat cycle conditions. The surface of treated limestone under heat cycles and water exposure appears smoother, resembling the surface of untreated off-spec limestone. Rust elements were also detected on limestone subjected to heat cycles. The smoother surface increased small particle content and reduced larger distinct particles in treated limestone under certain conditions, which is likely due to the detachment of sand particles during exposure cycles. A significant loss of sand particles was observed when the limestone was directly immersed in different solutions.

This suggests that prolonged exposure to harsh environmental conditions may weaken the integrity of the treated limestone by altering its surface texture and particle composition. These findings provide insight into the potential durability limitations of treated limestone in aggressive environments, highlighting the need for further investigation into long-term performance and practical applications in construction materials.

Using unsaturated polyester resin in treating off-spec limestone aggregates has significant environmental and economic implications. Environmentally, this method reduces the demand for newly quarried aggregates, thereby lowering carbon emissions and mitigating ecological damage from mining activities [32]. Experimental results indicate that resin treatment significantly enhances aggregate performance, reducing the ACV from 41% to 17%, which signifies improved compressive strength. Similarly, the AIV decreased from 11% to 5%, demonstrating increased resistance to impact loads.

Economically, the use of treated aggregates can lower construction and maintenance costs. Enhanced aggregate durability reduces road surface degradation, minimising long-term infrastructure expenditures [25]. However, the initial cost of resin treatment must be evaluated against the expenses associated with mining, transportation, and processing of conventional aggregates.

Despite these advantages, the environmental footprint of unsaturated polyester resin, a petroleum-based material, necessitates further investigation. A comprehensive life cycle assessment is crucial to determine whether the sustainability benefits of aggregate reuse outweigh the ecological costs of resin production and disposal. This study contributes to technical advancements in aggregate modification and sustainable innovation in the construction industry.

Therefore, the findings can be further elaborated through a comparative analysis, as shown in Table 6, between existing methods for enhancing aggregate performance and the new approach introduced in this study.

Table 6. Comparative analysis between existing methods and the new approach

Aspect	Existing Methods	New Method (Unsaturated Polyester Resin Treatment)
Types of Treatment	Mechanical, chemical, thermal, polymer/bitumen coatings	Surface treatment using unsaturated polyester resin
Main Objective	Improve strength, durability, and water absorption	Enhance strength, durability, microstructure, and environmental resistance
Crushing Strength (ACV Reduction)	Limited improvement; depends on method used [10]	Significant reduction in ACV (62.2%), improving strength
Impact Resistance (AIV)	Can be improved through chemical and thermal treatment, but is still affected by old mortar [13]	Improved impact resistance; AIV reduced to 5% from 11%
Abrasion Resistance (LA Test)	Polymer and bitumen coatings provide better wear resistance [26]	Improved abrasion resistance, reducing the LA abrasion value from 21% to 17%
Water Absorption	RCA and untreated aggregates have high absorption; chemical coatings help, but are not always effective [14]	Reduced water absorption significantly, meeting industry standards
Durability in Harsh Conditions	Some treatments degrade in extreme conditions like saltwater and acid exposure	Maintained better mechanical properties under NaCl, HCl, and thermal cycles
Microstructural Changes	Chemical and thermal treatments alter surface texture but may cause cracking [11]	SEM analysis showed a rougher surface, enhancing bonding and chemical interactions
Sustainability	RCA and recycled materials promote sustainability but may require additional processing [3]	Extends the use of local off-spec aggregates, reducing the need for new materials
Energy Consumption	Thermal and chemical treatments often require high energy input [28]	Low energy requirement since treatment is surface-based
Suitability for Road Construction	Some methods improve aggregates, but long-term stability remains a concern [29]	Demonstrated potential for use in road surfaces, meeting industry requirements

5. CONCLUSIONS

This study demonstrated that unsaturated polyester resin effectively enhances the mechanical properties of off-spec limestone aggregates, making them suitable for road construction. The treatment significantly reduced the ACV from 41% to 17% and the AIV from 11% to 5%, improving compressive strength and impact resistance. Additionally, LA abrasion values were lowered, enhancing aggregate durability.

SEM analysis revealed that resin-treated aggregates exhibited a rougher surface texture than untreated limestone. This improved surface morphology enhances mechanical interlocking between aggregates and cement paste, promoting better bonding and increasing concrete durability. However, prolonged exposure to environmental stressors, such as NaCl, HCl, and thermal cycles, led to partial surface degradation, indicating the need for further durability assessments.

From an environmental perspective, resin treatment supports sustainability by reducing reliance on newly quarried aggregates, thereby lowering carbon emissions and minimising ecological degradation. Utilising local off-spec aggregates decreases transportation-related environmental costs and aligns with circular economy principles by repurposing construction materials. However, as unsaturated polyester resin is a petroleum-based product, its long-term environmental impact, including production emissions and disposal concerns, requires further evaluation.

Economically, the enhanced durability of resin-treated aggregates can reduce road maintenance costs by extending pavement lifespan. While the initial cost of resin treatment must be considered, the potential savings in infrastructure repairs and material sourcing justify further exploration of its economic viability. Future research should conduct a comprehensive life cycle assessment and cost-benefit analysis to determine its feasibility on a larger scale, ensuring both environmental and economic sustainability in construction applications.

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