International Journal in Physical and Applied Sciences
Volume 11 Issue 10, October 2024 ISSN: 2394-5710 Impact Factor: 8.657
Journal Homepage: http://ijmr.net.in, Email: irjmss@gmail.com
Double-Blind Peer Reviewed Refereed Open Access International Journal

Comprehensive Evaluation of Chemical, Mineralogical, and Physical Properties of Cullet for Industrial Applications

Ibrahim Yaro Getso[a], Hassan U. J.[b], Fuad Musa[c] , Umar Salisu Ahmad[a] , Muhammad Sani Idris[a], Sani Garba Durumin-Iya[d]
[a]Department of Physics, Federal University of Education, PMB 3045, Kano State - Nigeria
[b]Department of Physics, Aliko Dangote University of Science and Technology, PMB3244, Kano State - Nigeria
[c]Science and Technical Schools Board, BUK Road Kano State - Nigeria
[d]Department of Physics, Faculty of Natural and Applied Science, Sule Lamido University, Kafin Hausa, Jigawa State Nigeria
Corresponding Author: igyaro779@gmail.com

ABSTRACT

Silicate – based wastes are produced in large quantities and have been considered for recycling and reuse. Among these, cullet – a significant silicate waste – makes up a substantial portion of urban waste. While much of it is recycled by the glass industry, a considerable amount still ends up in landfills. In recent decades, research has focused on the potential of cullet as an aggregate in the production of glass – ceramics, mortars, ceramics, cement, and concrete. This study examines the chemical, mineralogical, and physical properties of cullet for potential industrial applications. Cullet typically contains minerals such as SiO_2, Al_2O_3, and CaO in varying amounts. Analytical techniques like X – ray fluorescence (XRF), X – ray diffraction (XRD), and scanning electron microscopy (SEM) were employed to assess its surface morphology and internal structure. Results indicate that cullet is primarily composed of SiO_2, displaying a crystalline structure and a porous, cellular morphology with irregularly shaped particles. These findings suggest that cullet holds promise as a valuable material for various applications in the glass and cement industries.

Keywords: Cullet, Chemical Properties, Mineralogical Properties, SEM, DTA

INTRODUCTION

The recovery of container cullet for recycling often results in surplus quantities of the material, which offers opportunities for repurposing in various industries. Finely ground glass cullet, due to its pozzolanic properties, can be utilized as a cement component in concrete, enhancing its value and allowing the energy expended during glass production to be leveraged. One of the most common uses for broken glass is as an aggregate in road base applications or concrete [1-15]. This utilization is particularly promising because glass, being amorphous and rich in silicon and calcium, theoretically possesses pozzolanic characteristics. When finely ground, its cementitious nature has been demonstrated in study [2]. Incorporating glass into concrete also adds value by tapping into the energy retained from the glass manufacturing process.

However, the use of glass in concrete is not without challenges. Glass containers contain significant amounts of sodium and silicon, which can lead to alkali-silica reactions (ASR), a harmful chemical reaction that can cause expansion and cracking in concrete. Research has shown that when glass is used as a sand replacement in concrete, higher expansion rates are observed during ASR tests [16-20]. Fortunately, the addition of pozzolans can mitigate these issues and prevent the detrimental effects of ASR [21].

The use of mixed-colour glass cullet in cement-based materials offers an alternative recycling pathway to other uses such as abrasives, glass wool, or water filtration media. This application is promising due to the difficulties associated with recycling mixed-colour glass for new glass production. In cement-based materials, however, glass can exhibit two opposing behaviours: it can either cause damage due to alkali-silica reaction or benefit the concrete through pozzolanic activity. ASR is usually triggered by coarse particles of glass that contain amorphous silica. The breakdown of the silica network releases silica, which combines with alkali and calcium to form expansive gels, leading to the destruction of concrete. Several studies have explored the relationship between glass particle size and the expansion of mortars and concretes during ASR [15-22], although the results vary widely and are difficult to generalize. The critical particle sizes that trigger harmful expansion range from approximately 150 micrometres to over 1 millimetre, depending on factors such as glass type, colour, concrete composition, and testing conditions.

Globally, approximately 10 million tonnes of waste and crushed glass are produced in large cities, making up about 3-5% of all household waste. Since the 1960s, glass-asphalt mixtures have been explored as a method of recycling waste glass, and around 20 years ago, researchers began incorporating glass cullet into asphalt aggregate mixtures. The first applications of glass cullet in road pavements aimed to evaluate their resistance to moisture [10]. Experiments have shown that pavements containing waste glass reflect more light at night, which enhances road visibility and improves driving safety [11]. Additionally, using crushed glass in pavements has been found to improve vehicle braking and acceleration due to the increased internal friction and fracture strength provided by the glass particles.

Despite these benefits, the smooth surface of glass particles and their high silica content make them hydrophilic, increasing the pavement's susceptibility to water damage. This vulnerability can weaken the bond between bitumen and stone aggregates, resulting in pavement stripping. To prevent this, additives like hydrated lime are commonly used to maintain the beneficial properties of glass-asphalt mixtures while mitigating their weaknesses [12]. Glass particles also absorb less bitumen than traditional aggregates, resulting in lower bonding strength at the interface between glass and asphalt. This reduced bonding strength causes glass-asphalt mixtures to have lower overall fracture strength compared to conventional asphalt concretes. However, adding 1-2% lime to the mixture can counteract this deficiency, and higher percentages of lime can further improve the flexibility strength of the pavement [13-18].

Glass-asphalt mixtures containing 10-15% glass cullet by weight have been shown to perform well in surface coating mixtures. In addition to technical considerations, safety factors must also be addressed, such as the risk of skin cuts and tire punctures. To minimize these risks, the maximum allowable size of glass particles in pavements is typically set at 4.75 millimetres. In a study, 2% lime was added to the mixture as an anti-stripping agent, which improved the performance of pavements containing 10% waste glass compared to those without glass after one year of use [20-23]. Another recent study by [21], examined the dynamic characteristics of asphalt mixtures containing waste glass aggregates and found that glass-asphalt pavements had a higher stiffness modulus compared to conventional asphalt mixtures. The addition of 3% hydrated lime as an anti-stripping agent further increased the stiffness modulus of the glass-asphalt mixtures, demonstrating the effectiveness of lime in improving pavement durability.

Beyond its use in asphalt, cullet as a recycled material offers potential benefits in glass fabrication and other industries. Incorporating cullet into new glass products or using it to replace quartz in certain applications could enhance mechanical properties and reduce the environmental impact associated with waste glass. Replacing quartz with cullet in processes like sintering could affect variables such as sintering temperature, mold pressure, and soaking time. By studying the chemical and mineralogical properties of cullet, researchers can better understand the mechanisms that govern its strength and develop methods to further improve its mechanical properties.

This paper focuses on the characterization of cullet for industrial applications, particularly its use as a component in glass or cement. Techniques such as scanning electron microscopy (SEM), X-ray fluorescence (XRF), X-ray diffraction (XRD), and differential thermal analysis (DTA) were employed to examine the physical, chemical, and mineralogical properties of cullet. These analyses provide valuable insights into the suitability of cullet for use in various industries, including glassmaking and construction materials. Through a comprehensive understanding of its properties, cullet can be effectively utilized to enhance the performance and sustainability of products in these sectors, contributing to both economic and environmental benefits.

EXPERIMENTAL PROCEDURE

X – ray Fluorescence (XRF)

The sample underwent analysis using X-ray fluorescence (XRF). The instrument employed for this process was the Bruker S4 Pioneer XRF, operated at a voltage of 60 kV.

Scanning Electron Microscopy

The morphology of the sample was analysed using a JOEL-JSM-6380 instrument at the Mechanical Laboratory of Universiti Tun Hussein Onn Malaysia. A small amount of the sample powder was placed onto carbon tape attached to a holder, and excess powder was blown off with an air gun to ensure only fine particles remained on the tape. The sample was then placed in the SEM chamber for analysis. The scanning electron microscope (SEM) was operated at 10 kV, and an image of the sample was captured at a magnification of X100.

X – Ray Diffraction (XRD)

The samples underwent X-Ray Diffraction (XRD) analysis using an X-Ray Diffractometer to determine their silica structure. Before analysis, the samples were ground into a powder using a mortar and pestle due to their brittle nature. The powdered samples were then analysed with Cu Kα radiation, using a scanning rate of 0.05° per second, operating at 40 kV/20A, and a scanning speed of 0.05°/min, covering a 2θ range from 3° to 90°. The XRD analysis was performed using a Bruker D8 Advance diffractometer available at the Faculty of Civil Engineering.

Differential Thermal Analysis (DTA)

Thermogravimetric analysis (TGA) of the sample was conducted using a Lense in Thermobalance instrument at the Ceramics and Polymer Laboratory, UTHM. Understanding the thermal properties of the sample, particularly the melting point, is crucial for this study. Differential thermal analysis (DTA) provides insights into temperature changes, helping to determine the appropriate sintering temperature. For the test, a small amount of sample powder (11.3 mg) was heated and cooled at a rate of 10°C per minute, with the maximum temperature set to 1000°C.

RESULTS AND DISCUSSIONS

The SEM image of the cullet is shown in Figure 1 below. The particles exhibit a porous structure and have an irregular shape.

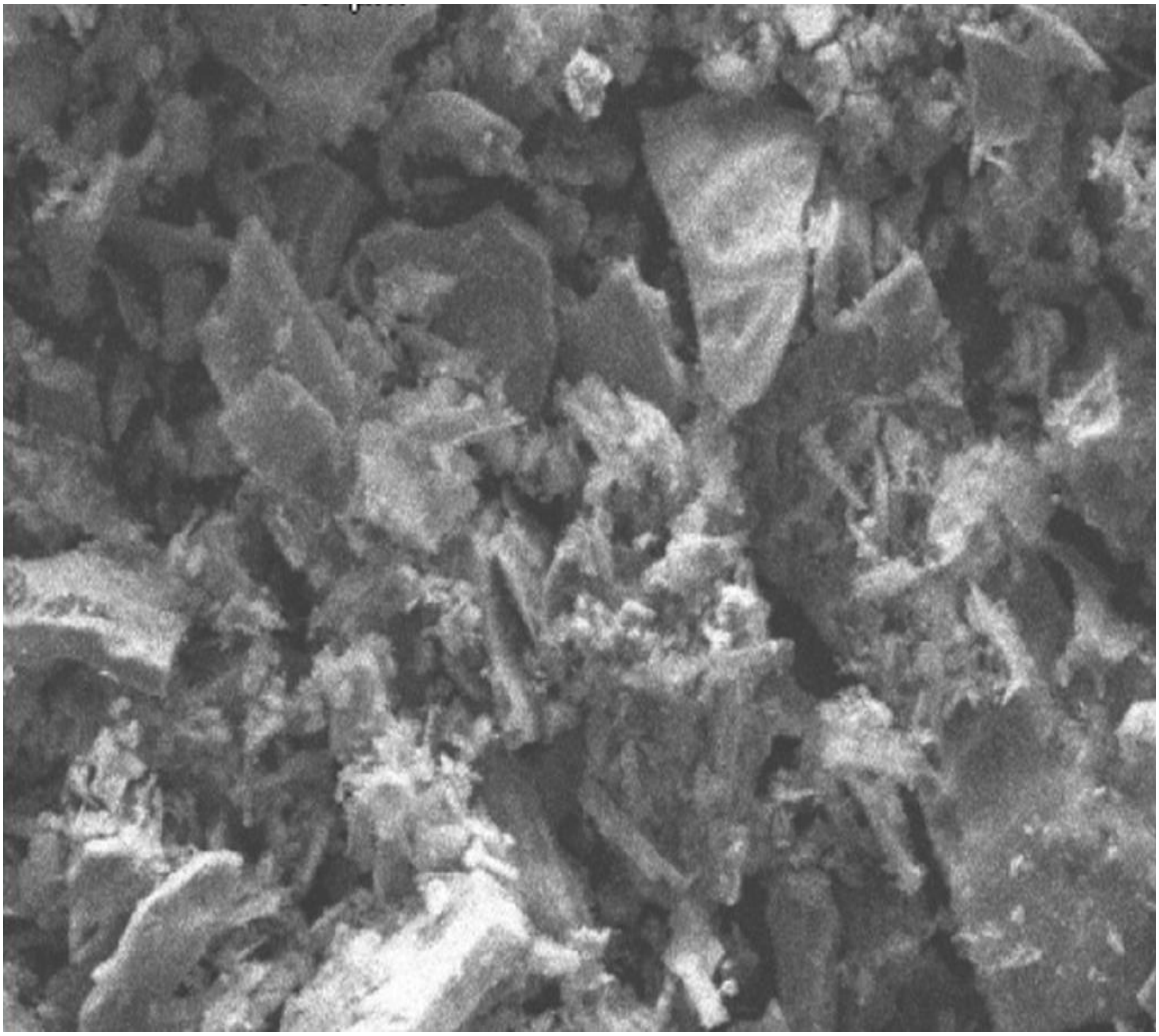

Figure 1: SEM of a Cullet (Magnification 1000X)

XRF of Cullet

The XRF analysis of the cullet is provided in Table 1. The cullet primarily contains SiO_2 (81.06%), with a lower concentration of fluxing agents (Fe_2O_3 + CaO + MgO + K_2O + Na_2O, totalling 15.45 wt%). The high content of SiO_2 and Al_2O_3 suggests the formation of a highly viscous liquid phase at elevated temperatures, which can trap gases. The fluxing agents, on the other hand, play a role in influencing the softening and melting points of the aggregates. Overall, the results indicate that cullet is a suitable material for industrial applications.

Table 1: XRF of Cullet

Chemical Constituents	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	LOI
Percentage Composition (%wt)	81.06	8.01	0.21	5.60	2.41	0.20	5.38	1.85	0.88

The XRD pattern in Figure 2 shows diffraction peaks at 2θ values of 20.990, 26.770, 36.620, 39.520, 42.490, 45.850, 50.230, 54.950, 60.100, 68.170, 72.430, 80.180, and 81.460. These peaks correspond to the characteristic signals of silicon oxide (SiO_2), as identified by the Joint Committee on Powder Diffraction Standards (JCPDS) file no. 27-1402. The sample consists of SiO_2 with a hexagonal lattice structure, space group P3121 (space number 152), a density of 2.71 g/cm³, and a unit cell volume of 113.20 m³.

The DTA graph of the cullet, displayed in Figure 3, shows an endothermic peak at 36.2°C, which is attributed to SiO_2, Al_2O_3, and CaO. Another endothermic peak at 100°C indicates the decomposition of SiO_2, while a separate endothermic peak points to the decomposition of Al_2O_3. During hydration, the intensity of the CaO peak diminishes, and the presence of hydrated products increases. The carbonation of Na_2O results in the presence of K_2O across all samples. At 699.4°C, the primarily amorphous hydration products are vulnerable to atmospheric CO_2. The XRD results confirm the presence of SiO_2 and Al_2O_3, with vitrified materials showing significant amounts of CaO.

Silicon serves as a network former in the glass structure, creating a highly cross-linked network of chemical bonds. Other elements, such as boron or germanium, can also act as alternative network formers in similar structures.

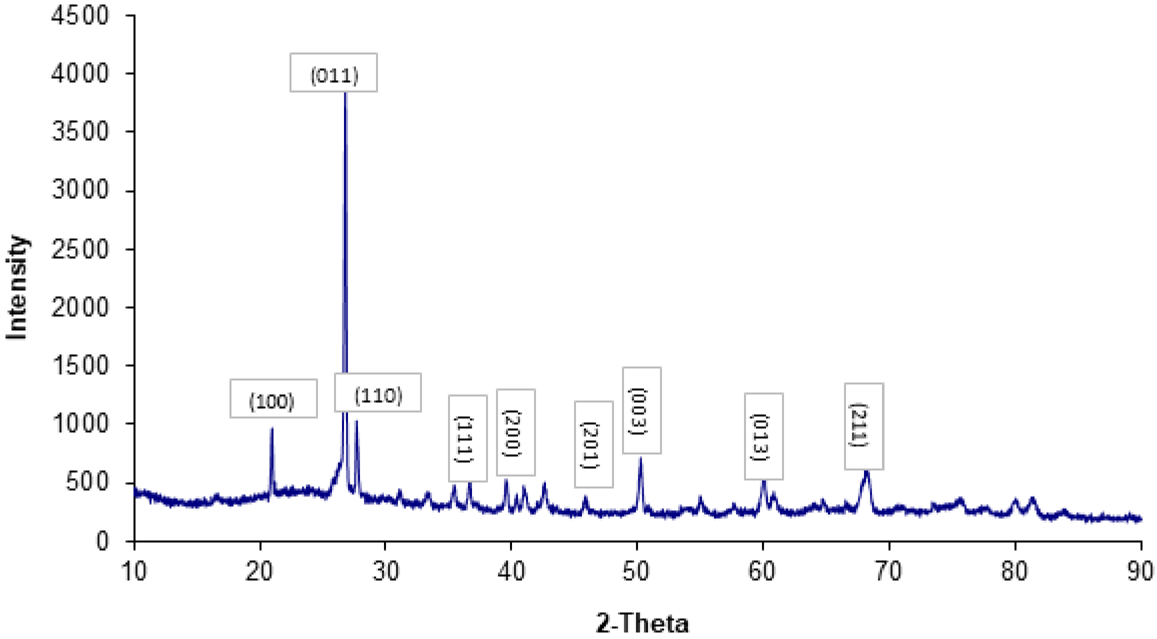

Figure 2: XRD of Cullet

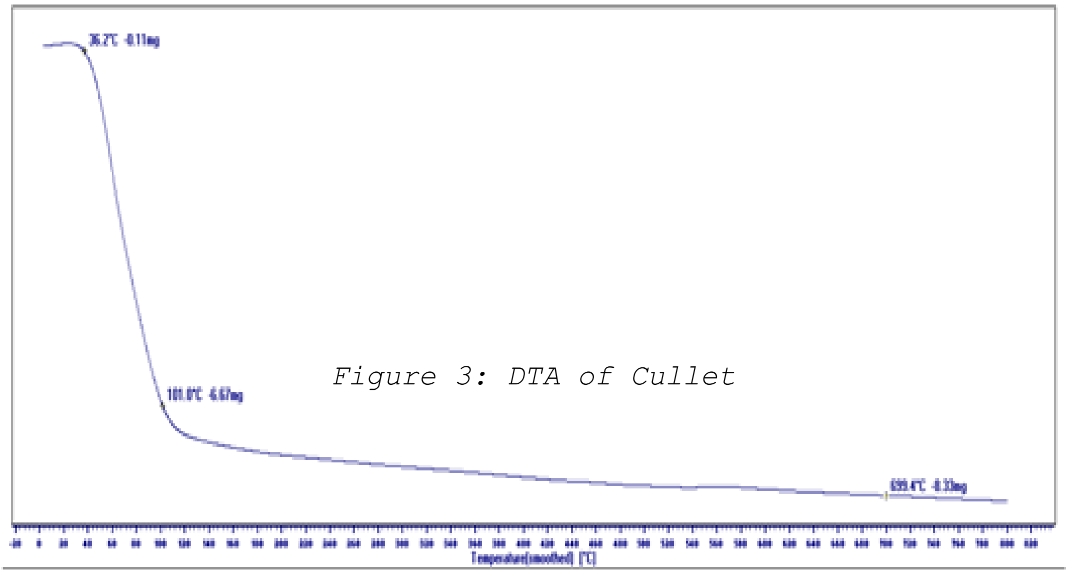

Figure 3: DTA of Cullet

The literature reveals numerous advantages associated with the incorporation of glass cullet into ceramic materials. These include reduced electrical energy consumption, improved material quality, conservation of ceramic raw materials, reduction in harmful emissions such as HF, and a decrease in waste disposal challenges [23]. However, one of the main obstacles to recycling discarded glass is the high cost of transportation to distant recycling facilities, making it economically unfeasible. Therefore, it becomes necessary to reuse or recycle the glass near its place of origin. Due to variations in the mineralogical and chemical composition of clays used in the ceramic industry, it is essential to assess the sintering performance of each type of clay when glass is incorporated.

The primary raw material for glass production is sand or quartz, which is the crystalline form of silica (SiO_2). This material consists of a continuous framework of SiO_4 (silicon-oxygen tetrahedra), with each oxygen atom shared between two tetrahedra. In amorphous glass structures, while the bonds within each silica tetrahedron are similar in length to those in the crystalline form, the arrangement of tetrahedra lacks long-range order, resulting in irregular three-dimensional distances.

In its purest form, glass can be made entirely from silica, known as "quartz glass" or "fused quartz." However, producing amorphous glass from pure silica is extremely energy-intensive, requiring temperatures of around 1900°C. As a result, quartz glass is typically reserved for applications demanding high chemical resistance, making it a specialized glass type.

To reduce the energy required for glass production, most glass consists of silica combined with other compounds. Silicon serves as the primary "network former," responsible for creating the highly cross-linked network of chemical bonds within the glass. Other elements, such as boron or germanium, can also act as alternative network formers. In addition to network formers, glass also contains "network modifiers," which are alkali oxides (e.g., sodium, potassium, lithium) added as fluxing agents to lower the melting point of the glass. Other modifiers include alkaline earth metal oxides (e.g., calcium, magnesium, barium, strontium) and various metal oxides, such as aluminium oxide.

These network modifiers alter the bonding relationships and structural groupings in the glass, leading to changes in its physical and chemical properties. The modifiers typically exist as ions, balanced by nearby non-bridging oxygen atoms that form a single covalent bond with the glass network and hold a negative charge to counteract the nearby positive ions.

CONCLUSION

The chemical, mineralogical, and physical properties of cullet have been thoroughly studied, revealing its potential as a valuable resource. Cullet is not only a rich source of SiO_2 but also contains significant amounts of Al_2O_3, CaO, and Na_2O, making it suitable for the production of a wide range of valuable products. The particles exhibit a porous structure with irregular shapes. With an SiO_2 content of 81.06% and a low concentration of fluxing agents (Fe_2O_3, CaO, MgO, K_2O, Na_2O totalling 15.45 wt%), cullet's composition ensures the formation of a highly viscous liquid phase at high temperatures, capable of entrapping gases.

The material's thermal properties are highlighted by an endothermic peak at 36.2°C, associated with SiO_2, Al_2O_3, and CaO, and another at 100°C, which can be linked to the decomposition of SiO_2, followed by a further peak related to Al_2O_3 decomposition. However, successful product development will require a deeper understanding of the chemical, mineralogical, and physical characteristics of cullet.

The data presented here offer valuable insights into the compositional and morphological attributes of both the surface and internal structure of cullet. Expanding the knowledge of its chemical composition, physical characteristics, and surface morphology is crucial for determining the suitability of cullet for various industrial applications.

Acknowledgement

The authors would like to acknowledge the financial support of Tertiary Education Trust Fund (TETFund) for sponsoring this research and Directorate for Research and Development Federal University of Education Kano, for facilitating the process of accessing the research grant.

References

[1] Henry, K. S., & Morin, S. H. (1997). Frost susceptibility of crushed glass used as construction aggregate. *Journal of cold regions engineering*, *11*(4), 326-333.

[2] Dyer, T. D., & Dhir, R. K. (2001). Chemical reactions of glass cullet used as cement component. *Journal of Materials in Civil Engineering*, *13*(6), 412-417.

[3] Idir, R., Cyr, M., & Tagnit-Hamou, A. (2011). Pozzolanic properties of fine and coarse color-mixed glass cullet. *Cement and Concrete Composites*, *33*(1), 19-29.

[4] Arabani, M. (2011). Effect of glass cullet on the improvement of the dynamic behaviour of asphalt concrete. *Construction and Building Materials*, *25*(3), 1181-1185.

[4] Shafabakhsh, G. H., & Sajed, Y. (2014). Investigation of dynamic behavior of hot mix asphalt containing waste materials; case study: Glass cullet. *Case Studies in Construction Materials*, *1*, 96-103.

Archibald, J. F., DeGagne, D. O., Lausch, P., & De Souza, E. M. (1996). Ground waste glass as a pozzolanic consolidation agent for mine backfill. In *International Journal of Rock Mechanics and Mining Sciences and Geomechanics Abstracts* (Vol. 6, No. 33, p. 271A).

[5] Polley, C., Cramer, S. M., & Cruz, R. V. D. L. (1998). Potential for using waste glass in Portland cement concrete. *Journal of materials in Civil Engineering*, *10*(4), 210-219.

[6] Shao, Y., Lefort, T., Moras, S., & Rodriguez, D. (2000). Studies on concrete containing ground waste glass. *cement and concrete research*, *30*(1), 91-100.

[7] Shi, C., Wu, Y., Riefler, C., & Wang, H. (2005). Characteristics and pozzolanic reactivity of glass powders. *Cement and Concrete Research*, *35*(5), 987-993.

[8] Shayan, A., & Xu, A. (2004). Value-added utilisation of waste glass in concrete. *Cement and concrete research*, *34*(1), 81-89.

[9] Shayan, A., & Xu, A. (2006). Performance of glass powder as a pozzolanic material in concrete: A field trial on concrete slabs. *Cement and concrete research*, *36*(3), 457-468.

[10] Wu, S., Yang, W., & Xue, Y. (2004). Preparation and properties of glass–asphalt concrete. *Wuham (China): Key Laboratory for Silicate Materials Science and Engineering of Ministry of Education, Wuham University of Technology*.

[11] Marti, M. M., & Mielke, A. (2002). *Synthesis of asphalt recycling in Minnesota* (No. MN/RC--2002-32,).

[12] Cyr, M., Rivard, P., & Labrecque, F. (2009). Reduction of ASR-expansion using powders ground from various sources of reactive aggregates. *Cement and Concrete Composites*, *31*(7), 438-446.

[13] Arabani, M. (2011). Effect of glass cullet on the improvement of the dynamic behaviour of asphalt concrete. *Construction and Building Materials*, *25*(3), 1181-1185.

[14]Day, D. E., & Schaffer, R. (1989). Glasphalt paving handbook. *Univ. of Missouri-Rolla, Rolla,*

Mo.

[15] Wenjiu, J. (2003). Recycle of waste glass. *Shanghai building materials*.

[16] Airey, G. D., Collop, A. C., & Thom, N. H. (2004, September). Mechanical performance of asphalt mixtures incorporating slag and glass secondary aggregates. In *Proceedings of the 8th Conference on Asphalt Pavements for Southern Africa (CAPSA'04), South Africa* (Vol. 12, p. 16).

[17] Nuhu, A. H. (2020). *Fabrication and characterizations of recycled aluminium-copper-cullet powder (AL-CU-CP) metal composite* (Doctoral dissertation, Universiti Tun Hussein Onn Malaysia).

[18] Purnawan, S. S., Manullang, R. J., & Wahyudi, K. (2020). Cullet In The Circular Economy Of The Glass Industry. *Indonesian Journal of Industrial Research*, *29*(2), 139-151.

[19] Powęzka, A., Ogrodnik, P., Szulej, J., & Pecio, M. (2021). Glass cullet as additive to new sustainable composites based on alumina binder. *Energies*, *14*(12), 3423.

[20] Duraman, S. B., & Li, Q. (2021, May). Recycled glass cullet as fine aggregate and partial cement replacement in concrete. In *IOP Conference Series: Materials Science and Engineering* (Vol. 1144, No. 1, p. 012010). IOP Publishing.

[21]Suleimenov, Z. T., Sagyndykov, A. A., & Abutalipov, E. A. (2024). Composite Glass-Crystal Facing Materials Based on Industrial Phosphoric Waste and Cullet. *Glass and Ceramics*, *80*(9), 392-395.

[22]Min'ko, N. I., Bessmertny, V. S., Zdorenko, N. M., Bondarenko, M. A., Isaenko, E. E., Tarasova, E. E., ... & Cherkasov, A. V. (2023). Environmental Aspects of Cullet Usage in Glass-Concrete Manufacturing. *Glass and Ceramics*, *80*(5), 233-240.

[23] Costa, F. B., Teixeira, S. R., Souza, A. E., & Santos, G. T. A. (2009). Recycling of glass cullet as aggregate for clays used to produce roof tiles. *Matéria (Rio de Janeiro)*, 1146-1153.

[23] Vieitez, E. R., Eder, P., Villanueva, A., & Saveyn, H. (2011). End-of-waste criteria for glass cullet: Technical proposals. *JRC Scientific and technical reports*.