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Waste glass as a supplementary cementitious material in concrete – Critical review of treatment methods

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ABSTRACT

Concrete is the most widely used construction material and, for the most part, is produced using non-renewable natural resources and energy intensive processes which emit greenhouse gasses. There exists an opportunity to improve the sustainability of this industry by further exploring the use of alternative materials. As an aggregate, glass bottle waste has faced specific challenges including bond, ASR gel production, and strength degradation of the concrete. This paper reviews the literature pertaining to incorporating waste glass into concrete as a supplementary cementing material. Pozzolanic properties of waste glass as an SCM and ASR are related to particle size and percent addition. Lithium additives control ASR expansion; however, the mechanism of this control has yet to be defined.

1. Introduction

The concept of utilizing waste material for building applications has a long and successful history, which includes fly ash, slag, and silica fume. These once problematic, land filled waste materials are now considered valuable commodities for use in enhancing certain properties of concrete. A waste material which continues to face challenges as a component of concrete is bottle glass. The benefits of developing alternative or supplementary cementing materials as partial replacements for ordinary Portland cement (OPC) powder are described by Malhotra and Mehta [1], who divide them into ecological, economic, and engineering categories.

Ecological or environmental benefits of alternative materials include (1) the diversion of non-recycled waste from landfills for useful applications, (2) the reduction in the negative effects of producing cement powder, namely the consumption of non-renewable natural resources, (3) the reduction in the use of energy for cement production and (4) the corresponding emission of greenhouse gasses. The economic benefits of using alternative materials are best realized in situations where the cost of the alternative material is less than that of cement powder while providing comparable performance. This cost must consider the source of the alternative material, its transportation, processing, and should consider savings through diversion, such as tipping fees and landfill management costs. The engineering or technical benefits of alternative materials are realized when a specialized use for such material may be developed, such that the use of the alternative material is more desirable than use of concrete made with OPC alone.

There exists an opportunity to realize these potential benefits even in communities where recycling programs are in place. Although a curb side recycling program has meant increasing rates of recycling in Ontario since 1987, only 68% of alcohol containers, for example, are recovered through recycling programs, and much of this material is broken and of mixed colour, making high value recycling difficult and impractical. As a result, waste glass can often end up in landfills [2]. Alcohol containers alone make up 44% of curb side recycled material, and by 2015, these containers are predicted to contribute 116,000 tonnes of material to the collection stream [3].

Early attempts to incorporate waste glass in concrete tended to focus on aggregate replacement. The properties of glass seemed comparable to those of large aggregate in terms of constitution, strength and durability, and the larger size of the glass meant lower processing costs. These early attempts however, were unsuccessful due to the alkali–silica reaction (ASR) which takes place in the presence of the amorphous waste glass and concrete pore solution. In addition to ASR, several other challenges exist in incorporating waste glass into concrete. Using waste bottle glass limits the size and shape of particles for coarse aggregate. The maximum size of one dimension of the aggregate will always be the thickness of the bottle. Crushed waste bottles will therefore tend to form large aggregate which is flat and elongated, and may negatively affect workability [4], and which has been attributed to observed

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decreases in compressive strength [5]. Waste glass also presents a challenge in terms of contamination and consistency in the waste stream. Most waste glass that cannot be recycled is broken and of mixed colour, which will cause variation in the chemical composition of the waste. There may be contamination due to paper and plastic labels, caps and corks, and sugars remaining from the original use and content of the bottles [6]. Moreover, seasonal fluctuation in the waste stream would be expected.

In order to overcome the limitations of ASR, several treatment methods were explored, which generally included mechanical and chemical treatments. In exploring these methods, key developments have been made. Experimentation using mechanical treatments looked at reducing the particle size of the waste glass through grinding. The results of this work indicated that as the particle size of the waste glass was decreased, the tendency for ASR formation also decreased. Furthermore, the results yielded an increase in the compressive strength of concrete made with very fine waste glass [7,8]. Chemical treatments were initially explored in detail by McCoy and Caldwell [9], where a series of salts, acids, organic and proprietary compounds, and proteins were used in mortar bars containing Pyrex glass in an effort to identify which materials may reduce ASR. This series of tests indicated that the use of lithium compounds significantly reduced ASR related expansion.

This paper provides a critical review of research initiatives reported in the literature, the aim of which is to improve the feasibility of incorporating waste glass into concrete through various treatments. The review is weighted on the progress made to develop waste glass into a viable Supplementary Cementing Material (SCM). The focus of waste glass utilization has shifted in more recent years from aggregate to mineral additive in light of the observations related to particle size.

2. Challenge of ASR in the use of waste glass

Use of waste glass in concrete first focused on aggregate replacement. Phillips and Cahn [6] attempted to introduce waste glass as a partial replacement of the fine and coarse aggregate in concrete masonry block, the production and use of which is less conducive to ASR gel production, and which allow early age monitoring. Using amber glass in a high-alkali, fine cement, tested properties of the blocks generally fell near or within accepted limits, although strength gain was slow and below typical at 28 days. Phillips and Cahn observed that the main challenges in utilizing waste glass would be the removal of contamination, processing, and cost. Johnston [4] also considered waste glass as a coarse aggregate, and found that only by using low alkali cement or high percentages of pozzolans such as fly ash could satisfactory strength and expansion performance be achieved to one year.

The obvious challenges encountered during this preliminary work deterred interest in the idea of alternative uses for waste glass in the concrete industry for many years. In the last decade of the 20th century, a resurgence in interest has developed as a result of increased awareness of several social and global elements, including greenhouse gas emissions and energy costs.

Experimentation with the addition of waste glass in concrete has been closely related to the study of alkali–silica reactivity, where production of ASR gel in the presence of reactive aggregates causes damaging expansion in concrete. The topic of ASR reactivity has been thoroughly covered in the past 30 years. In this discussion, it is important to impart only that the mechanism by which a reactive aggregate can form ASR gel, leading to expansion and ultimately causing damage to concrete, is dependent on the presence of amorphous silica, which is the major component of waste glass, the presence of alkali hydroxides in pore solution, and cement reaction products [10]. An early study by Diamond and Thaulow aimed to determine the relationship between ASR production and reactive particle size, revealed that there is no limiting size for reaction. However, they observed that the reaction rate is greater at finer particle sizes [11]. Work by Shayan and Xu [7], as well as Jin [8], indicated that a decrease in particle size leads to a pessimum effect in terms of ASR expansion which was related to an increase in reactivity. Jin et al. [12] further hypothesized that the pessimum observed is related to a competition between ASR rate of reaction and rate of transport of ASR gel away from the reaction site, where at a certain particle size, the reaction rate is great enough and the transport rate low enough that a maximum expansion is observed. Fig. 1 shows some of these results, where depending on the glass composition and environment, a maximum ASR expansion tends to occur at an intermediate value. For most cases, both the local maximum and minimum expansion occurs at a very small particle size, typically less than 100 μm [11–14].

Effectiveness of methods such as chemical treatment and surface abrasion in improving the performance of concrete containing waste glass through prevention of expansive behaviour, and the effect of different glass colours on ASR expansion was reported by Meyer [15]. Polley et al. observed a pessimum effect with respect to the percentage of waste glass added to the concrete as a replacement of fine aggregate combined with fly ash [16]. A pessimum percentage was also observed by Kawamura and Fuwa [17], but not by Jin et al. [12].

Both the degree of reaction and the nature of the products are known to influence the ASR damage associated with the use of waste glass. Many attempts have been made to predict the swelling behaviour of the gel based on either the chemical composition of the initial reactants or the reaction product itself. Although a relationship has been observed between the ratio of alkali and silica in the gel and the resulting swellability [18–20], the relationship is too complex to allow accurate prediction due to localized variation in chemical composition of the gel.

3. Waste glass as a pozzolan

One of the limiting factors in the use of alternative materials as pozzolans in concrete is the lower reactivity of the materials when compared to cement powder. Overcoming this limitation requires practical methods for increasing the reactivity of SCMs. Shi and Day [21] effectively used salts as chemical activators to increase the reactivity of natural pozzolans, as well as several other methods, including mechanical and thermal treatments [22].

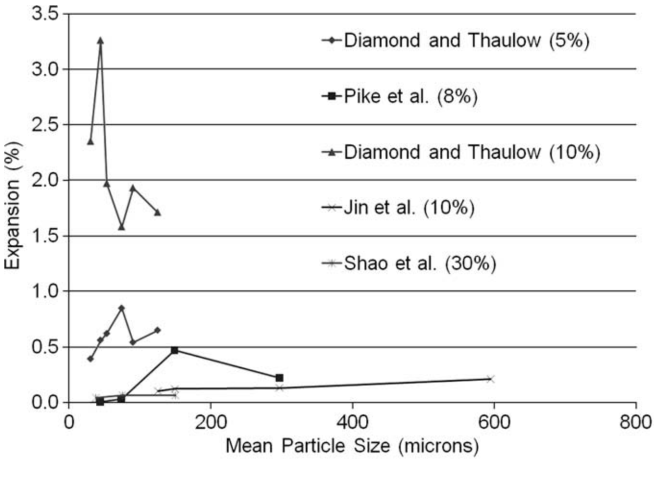

Fig. 1. Expansion vs. glass particle size for various percent replacements [11–14].

A comparison of the methods by Shi and Day indicated that the most effective method for developing reactivity in natural pozzolans was chemical activation, which improved both the initial reaction rate and the final strength. The reactivity of the treated pozzolanic material was measured in terms of the compressive strength and total hydration of the material.

The chemical compositions of some of the pozzolans are compared to the composition of waste glass in Table 1. With the exception of Al_2O_3 and CaO, the percentages of the main constituents are similar [21,23–26]. It is reasonable to expect that some success may be achieved by applying similar treatment to waste glass in order to improve its pozzolanic properties. The properties which influence the pozzolanic behaviour of waste glass, and most pozzolans in general, are fineness, composition, and the pore solution present for reaction. Based on observed compressive strengths, Meyer et al. [27] postulated that below 45 μm, glass may become pozzolanic. The pozzolanic properties of glass are first notable at particle sizes below approximately 300 μm. Below 100 μm, glass can have a pozzolanic reactivity which is greater than that of fly ash at low percent cement replacement levels and after 90 days of curing [26,28].

The pozzolanic reactivity of fine waste glass is observed as an increase in compressive strength, which can be seen in Fig. 2. In the reported data [26,14], compressive strength is highest for specimens containing very fine glass (<100 μm), and the strength decreases as particle size increases. Fig. 3 [7,14,29–33] shows the effect of percentages of waste glass replacing OPC and fine aggregate on the compressive strength of mortar bars. The results show that a cement replacement between 10% and 20% yields the highest strength, while fine aggregate replacement of up to 40% has little effect on compressive strength.

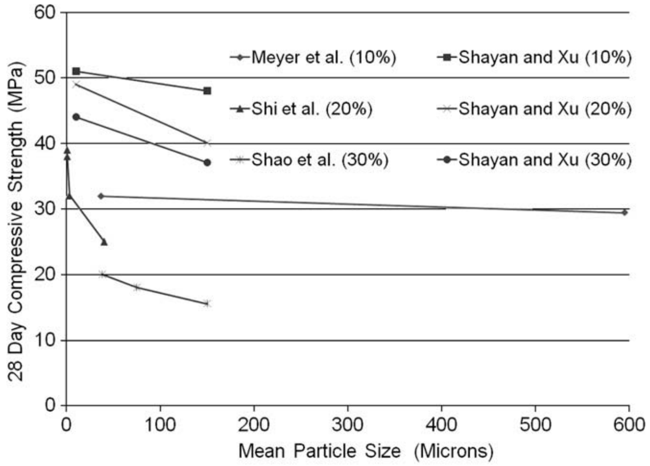

Fig. 2. 28-Day compressive strength vs. glass particle size for various percent replacements [14,26].

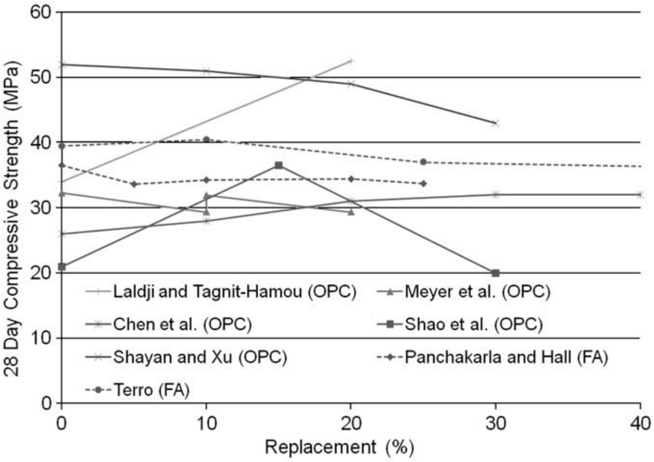

Fig. 3. 28-Day compressive strength for various percentages of waste glass replacing OPC and fine aggregate (FA) [7,14,29–33].

4. Lithium treatments

The use of lithium compounds has been explored for ASR mitigation as well as promoting pozzolanic reactivity. Some of the compounds which have been explored include lithium carbonate (Li_2CO_3) [34,17] lithium hydroxide (LiOH) [35,36], lithium nitrate ($LiNO_3$) [37,38], lithium chloride (LiCl) [39] and lithium fluoride (LiF) [40]. Lithium is unique among the alkali metals because it has the smallest atomic radius to possess the single valence electron in this group, yet a comparatively large hydrated radius and polarizing power. The properties of concrete and mortar specimens containing waste glass treated with lithium are also unique. Using lithium to stabilize dissolved silica, Lawrence and Vivian [36] were able to demonstrate that silica was slower to dissolve in the presence of lithium, and the product had different properties than that of other alkalis. Confirmed using ^{29}Si-NMR by Wijnen et al. [41], this low dissolution rate was accompanied by a lack of repolymerization of the silica species and the presence of a microcrystalline lithium silicate precipitate (Li_2SiO_3). Both silica dissolution and lack of repolymerization, as well as aggregation of particles, were observed by Kurtis et al. [39,42] using imaging techniques. While several authors have observed a decrease in dissolved silica in the presence of various lithium compounds, using LiOH specifically can result in an increase in dissolved Si while still mitigating ASR expansion [37,43,44]. The lack of repolymerization and the formation of aggregated or crystallized products are hypothesized to be related to the lack of ASR expansion in the presence of lithium. Fig. 4 shows expansion measured over an extended time frame as it relates to the Li/Na ratio of the product for various lithium treatments [35,45,46]. For most cases, there is a minimum

Table 1
Chemical composition of waste glass and other pozzolans.

Compound	Waste glass [23]	Volcanic ash [21]	Volcanic pumice [21]	Slag [24]	Silica fume [25]	High Ca fly ash [25]	OPC [26]
SiO_2	63.79	73.68	65.74	35	90.9	39.21	20.33
Al_2O_3	3.02	12.25	16.72	12	1.12	16.22	4.65
Fe_2O_3	1.57	2.2	3.58	1	1.46	6.58	3.04
CaO	13.01	1.13	3.33	40	0.69	22.78	61.78
MgO	0.89	0.23	0.95	–	0.77	2.35	3.29
K_2O	0.54	3.95	3.05	–	–	–	0.59
Na_2O	11.72	3.6	4.48	0.3	–	–	0.24
SO_3	0.165	0.32	0.65	9	0.38	4.3	3.63
LOI	4.55	3.05	2.4	1	3	2.1	–

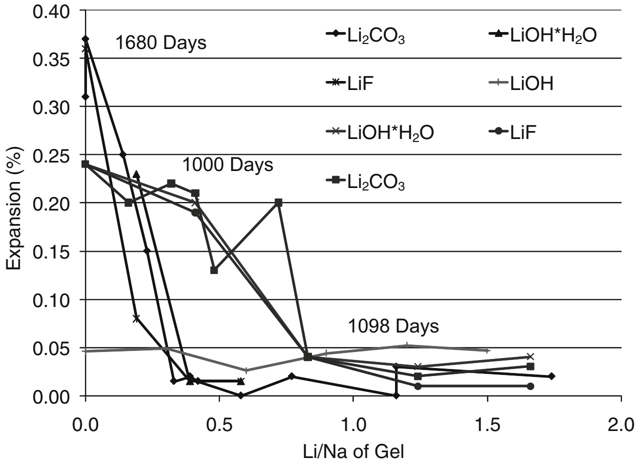

Fig. 4. Li/Na of gel vs. measured expansion after 1000, 1098, and 1680 days [35,45,46].

expansion observed at an intermediate value of Li/Na, however; depending on the reactive aggregate used, expansion may still exceed acceptable limits [46].

Work by Ramyar [40] attempted to relate the Ca/Si ratio of products in the presence of lithium compounds to the resulting structure, where two distinct products, massive and crystalline, were observed. However, no clear relationship could be determined. A decrease in CaO/SiO_2 was related to larger expansion with the exception of LiF as an additive. Further observations of the product of ASR reactions containing lithium by Kawamura and Fuwa [17] show that the CaO/SiO_2 ratio decreases and becomes more consistent throughout the sample as the lithium content is increased, and the resulting expansion is decreased, most notably for dosages of lithium greater than 0.5%. This result has in turn been related to the ratio of lithium to total alkali, where a threshold ratio to control expansion has been identified as ranging between 0.6 and 0.9, depending on the experimental program and the material tested [47]. There is a narrow range for the ideal concentration of lithium in the system, which may indicate a pessimum due to the additional OH^- liberated by the lithium compound [37,44,48]. Thomas et al. have observed that outside of the ideal lithium content, increased expansion may actually occur when compared to the control specimen [49]. Primary challenges still facing the use of lithium treatments include the absorption of large amounts of added lithium into the early hydration product, which, once absorbed, may not be available in later life, the increase in OH^- concentration [37], and the concerns of safety when working with reactive materials.

5. Waste glass as an SCM

The use of any supplementary cementing material will depend on its performance, namely its strength, durability, and volumetric stability over time. In the case of waste glass, this performance is further challenged by the tendency for ASR to occur. The controlling process between a beneficial pozzolanic reaction, which would improve the performance of glass as an SCM, and a damaging ASR is the production of either a stable or swelling product. Depending on several factors, including calcium content, particle size, and alkalinity, the dissolved silica will repolymerize into expansive gel, hydrate into C–S–H, or a combination of both [48]. A theoretical mechanism by Urhan [50] suggests that at any time in a concrete containing either reactive aggregate or pozzolanic material, ASR gel and C–S–H, as well as a range of intermediate products, can be found. According to this mechanism, availability of calcium ions in combination with a relatively high rate of C–S–H formation will favour the pozzolanic reaction, and over time, any ASR product will take on the texture of C–S–H. When reacting as a pozzolan, glass powder will follow Eq. (1) [50], and the product will be a type of calcium silicate hydrate.

Table 2
Pozzolans from Table 1 as SCMs, qualification under ASTM standards.

ASTM C 618-05 requirements [52]		Waste glass	Slag	Silica fume	Fly ash
$SiO_2 + Al_2O_3 + Fe_2O_3$, min%	70	68.4	48.0	93.5	62.0
SO_3, max%	4	0.2	9.0	0.4	4.3
Moisture Content, max %	3	–	–	–	–
LOI, max%	10	4.6	1.0	3.0	2.1

$$SiO_2(s) + Ca^{2+}(aq) + 2OH^-(aq) \rightarrow n_1CaO \cdot SiO_2 \cdot n_2H_2O(s) \tag{1}$$

When the reaction of glass results in ASR gel, the chemical equation is similar, however, sodium, potassium, or other alkalis may be substituted for calcium as shown by Eq. (2) [18].

$$SiO_2 + 2Na^{2+}(K^+) + 2OH^- \rightarrow Na_2(K_2)SiO_3 \cdot H_2O \tag{2}$$

The final product is more likely similar to a precipitate of the composition given in Eq. (3), existing within a sol/gel matrix of calcium silicate hydrate with a Na_2O/SiO_2 ratio near 0.19.

$$0.16Na_2O \cdot 1.4CaO \cdot SiO_2 \cdot XH_2O \tag{3}$$

This system may be the cause of variability in the swelling properties of ASR products [51].

Wang and Gillott [52], as well as Jin [8] have suggested that the mechanism which differentiates between pozzolanic and ASR products could be simply the degree of aggregation or particle size of the silica source. They postulated that the two reactions will compete with one another and the ease with which calcium ions can reach the pozzolanic reaction leads to a more stable and less expansive product. The question remains as to the specific role of lithium within the reaction product and its observed ability to control expansion in the system for various particle sizes and rates of reaction. In addition, the ability of LiOH to increase pozzolanic reactivity while decreasing ASR in the presence of glass or reactive aggregates seems contradictory.

The use of waste glass as a supplementary cementing material has demonstrated mixed results over a range of concentrations, particle sizes, and treatments, as was shown in Fig. 2 and Fig. 3. According to the requirements of ASTM standards [53] for the use of natural pozzolans, Table 2, glass has the potential to acceptably function as an SCM. However, proper methods must be developed to control the ASR/pozzolanic reaction and influence a non-destructive, non-swelling product. The form of this product has not been identified. It may be a lithium silicate, or perhaps a pozzolanic form of C–S–H, which has the potential to contribute additional strength.

6. Concluding remarks

The success, or lack thereof, in utilizing waste glass in concrete can be attributed to observed ASR damage. Although the collective understanding of ASR has improved dramatically through extensive research, it is still not possible to predict damage based on either the composition of the reactants or the reaction product. This prevents a fine level of control over the reaction.

As researchers moved from waste glass as an aggregate to waste glass as a replacement for OPC, pozzolanic properties were observed at particle sizes below 300 μm. These pozzolanic properties, however, are relatively weak, especially at larger particle sizes, and require enhancement. One possible form of enhancement used for natural pozzolans was chemical treatment.

The similarity between the ASR and pozzolanic reactions observed for waste glass in concrete suggests that they are closely related and may be simply various stages of one another subject to several controlling factors, including particle size, pore solution, and chemical composition.

Treatments with lithium appear to control the reaction by limiting the ASR expansion; however, the application of these treatments is not yet understood. There is still a very clear gap in understanding the exact mechanism at work and the role of lithium compounds in the mechanism of control.

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