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GROUND WASTE GLASS AS A SUPPLEMENTARY CEMENTITIOUS MATERIAL

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The cement industries worldwide are currently trying to find alternative cementitious materials for reducing the environmental impact and promoting sustainability. Among them, one is waste glass utilization. A major effort has been made to develop waste glass as aggregates in the concrete industry without complicated procedure. Applications of waste glass to the concrete aggregates are partly limited and often not successful due to the problem of alkali-silica reaction (ASR) between a larger amount of alkali and reactive silica in the waste glass resulted in severely aggravating durability of concrete. Possible method of reusing waste glass has been also developed on the utilization as supplementary cementitious material (SCM) including the economic and environmental concerns of waste glass utilization, if it is ground small enough, with suppressing the ASR expansion by pozzolanic activity of amorphous silica in waste glass powder. Many results of previous researchers as SCM of grounded waste glass are evaluated in this review.

Keywords: cement, waste glass, reactive silica, pozzolanic, concrete, aggregates

ИЗМЕЛЬЧЕННЫЕ СТЕКЛООТХОДЫ КАК ДОБАВКА К ВЯЖУЩИМ МАТЕРИАЛАМ

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Цементная промышленность во всем мире в настоящее время пытается найти альтернативные добавки к цементным материалам для снижения воздействия на окружающую среду и обеспечения устойчивости. Среди них важное направление – утилизация отходов стекла. Были предприняты значительные усилия по разработке методов использования отходов стекла в качестве добавок в бетонной промышленности без сложных дополнительных процедур. Применение отходов стекла в качестве бетонного заполнителя частично ограничено и часто не приводит к успеху из-за щелочно-кремнеземной реакции (ЩКР) между большим количеством щелочи и реактивным кремнеземом в измельченном стеклоотходе, что приводит к серьезному ухудшению долговечности бетона. Разработан также возможный способ повторного использования стеклобоя в качестве дополнительного вяжущего материала (ВВМ) с учетом экономических и экологических соображений утилизации стеклобоя, если отход из стекла достаточно мелко помола, с подавлением реакции ЩКР за счет пуццолановой активности аморфного кремнезема в порошке из отходов стекла. В этом обзоре оцениваются многие результаты предыдущих исследователей по использованию измельченного стеклобоя в качестве добавки к цементу.

Ключевые слова: цемент, стеклобой, реактивный кремнезем, пуццолан, бетон, заполнители

BOG'LOVCHILARGA QO'SHIMCHA SIFATIDA MAYDALANGAN SHISHA CHIQUINDILARIDAN FOYDALANISH

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Dunyo bo'yicha sement sanoati hozirgi kunda atrof-muhitga salbiy ta'sirni kamaytirish va barqarorlikni ta'minlash uchun sement materiallariga muqobil qo'shimchalarni o'rganishga bag'ishlangan ishlar amalga oshirilmoqda. Ular orasida shisha chiqindilarini qayta ishlash muhim yo'nalishlardan biri hisoblanadi. Chiqindilarni beton sanoatida qo'shimchalar sifatida murakkab jarayonlarsiz ishlatisht usullari o'rganilmoqda. Shisha chiqindisidan betonga qo'shimcha sifatida foydalanish qisman cheklangan va ko'pincha ishqoriy-kremniy reaksiyasi (IKR) tufayli ishqoriy muhit va reaktiv kremniyning yuqori miqdori o'rtasidagi ishqoriy shisha chiqindilari, bu esa betonning chidamliligini jiddiy ravishda yomonlashishiga olib keladi. Agar shisha chiqindilari etarlicha mayda maydalangan bo'lsa, shisha materialni qayta ishlashning iqtisodiy va ekologik jihatlarini hisobga olgan holda, qo'shimcha biriktiruvchi material (ACM) sifatida bog'lovchiga qo'shish mumkin. Shisha chiqindilaridan olingan kukun tarkibidagi amorf kremniyning puzolan faolligi yuqori bo'lib, ushbu sharhda sementga qo'shimcha sifatida maydalangan shisha chiqindilaridan foydalanish bo'yicha oldingi ilmiy ishlarni natijalari keltirilgan va baholangan.

Kalit so'zlar: sement, shisha chiqindisi, reaktiv kremniy, silika, puzolan, beton, agregatlar

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Introduction

Cement production is an energy-intensive and responsible for the larger portion of global man-made CO₂ emissions. The CO₂ emission mainly comes from calcination of limestone in the raw materials and fuel combustion in the kiln process for the cement production. Hence, nowadays so many diversified efforts have been executed by the utilization of economical, ecological or environmental benefits of alternative materials in cement industry including as [1, 2]: (i) the diversion of non-recycled waste for useful applications, (ii)

the reduction in the consumption of non-renewable natural resources, (iii) the reduction in the use of energy for cement production, and (iv) the reduction in the emission of greenhouse gases. The economic benefits of using alternative materials are already realized in situations where the cost of the alternative materials is less than that of existing materials in the cement production providing comparable performance of cement paste or concrete. One of the most effective ways to reduce economically the cement manufacturing cost as well as environmentally CO₂ emission in the cement in-

dustry is to partially substitute cement by SCMs, such as granulated blast furnace slag or fly ash, etc.

The engineering or technical advantages of these SCMs are already realized, developed, and expanded to the more desirable and useful construction areas than those of ordinary Portland cement (OPC) concrete alone. Since these SCMs are generally by-products of different industries, utilization of SCMs not only reduces consumed energy and resources, and CO₂ emission, but also prevents industrial by-products from accumulating in landfills. In addition, SCMs improve fresh and hardened concrete properties with ecological benefits. Consequently, most of the cement industries worldwide nowadays are expanding to use blended cements or additives in concrete by the cementitious materials of fly ash and/or blast furnace slag, with their own material and testing standards.

These SCMs are partly limited to utilize depending on the country's industry situation. In particular, in most of the developing countries where even cement demands are greatly increasing, they could not use or expand the amount of fly ash and/or granulated blast furnace slag in cement industry due to the limited or not-owned industry situation, such as coal-fired power plants and/or blast furnaces of iron and steel plants. Hence, sustainable SCMs appropriating to the country's industrial situation should be focused, found and developed for economically and environmentally balanced cement industry to satisfy the cement demand. For example, since coal-fired plants are very few and no blast furnace is present in Uzbekistan, even if they are imported from near countries, costly and quantitatively, it may be not enough to use as SCMs. As a result, SCMs suited to its own circumstance should be developed and expanded to meet the demand of building industry.

The main chemical compositions of widely used SCMs are generally aluminosilicate (SiO₂-Al₂O₃) or calcium aluminosilicate (CaO-SiO₂-Al₂O₃). By the economically and environmentally pushing cement industry and higher demand of cement consumer, the R&D activities in academia and industry are very active to explore more alternative SCMs from a practical point of view. Among them, one is waste glass.

Glass is usually a transparent, non-crystalline amorphous solid that has many important applications in a variety of industries worldwide. The most common type of glass is made primarily with soda ash (sodium carbonate, Na₂CO₃) and silica, as well as other additives at high temperature about 1400~1600 °C followed by cooling where solidification occurs without crystallization. Recycled glass (cullet) is also often used in the production of new glass. The continu-

ous consumption of inorganic materials instead of organic ones by lifestyle improvement leads to an increased amount of generation of solid waste. Most commercial glasses can be categorized into six types as: fused silica, soda-lime, borosilicate, aluminosilicate, barium, and lead glasses. Small amounts of additives are often added during the production of glasses to give glasses with different colors or improving specific properties. Fused silica glass is almost consisted about 100% SiO₂, and typical chemical composition of other glasses except small amount of additives used for color purpose is shown in Table 1 [3].

Table 1

Typical chemical composition of selected glass types

Element	Type of glass				
	Soda-lime	Boro-silicate	Alumo-silicate	Barium	Lead
SiO ₂	66~75	72~81	57~64.5	36~65	32~63
Al ₂ O ₃	0.5~1.5	1~6	16~24.5	2~4	0~2
CaO	5~12		8~10	~2	
MgO	0.1~4		7~10.5	~2	
Na ₂ O	12~17	4~7	0.5~1	~7	1~8
K ₂ O	0~3	~1		~9	2~9
B ₂ O ₃		11~15	4~5	~10	
BaO				~2	
PbO				2~41	22~65

A world glass industry produced a volume of 140 Mt in 2016 compared to 115 Mt in 2007 [4, 5]. Glass is an example of a resource that has unlimited recycling potential and can be, theoretically and completely, recycled with virtually no loss of physical quality. However, due to the complicated procedure of cleaning, separating, and sorting, unlimited recycling is not an easy task and could be restricted due to the variation of chemical composition and mixed color glass above all [6, 7]. Although waste glass is well collected, different color glass is often intermixed. Mixed color glass is hardly recycled because a mixing of coloring agents results in an unpredictable and uncontrollable color in the new glass of the manufacturing process. Also, the varying chemical composition of glass is the main reason why most waste glass cannot be remanufactured into glass products [6]. Even recycled ratio of waste glass is greatly different according the countries worldwide, on a global scale, this only amounts to a recycling rate of less than 35% [8]. Unrecycled waste glasses occupy huge parts of the landfills spaces, and the nonbiodegradable nature of glass causes environmental pollutions [9].

With faced with this situation, a major effort has been made to develop waste glass as a construction material as a mixed waste glass without complicated procedure. Above all, the use of

waste glass as aggregate for concrete has been attempted decades ago, and many results in the concrete application on aggregate replacement have been obtained on the operation, strength and durability of concrete [10-14]. Using waste glass limits the size and shape of particles for aggregates, because the shapes of flat, elongate, smoothness, sharp edge and harsh textures of crushed waste glass harmed the fluidity of concrete, and consequently negatively affects workability [11-13] and simultaneously decreases compressive strength due to the low adhesion between the smooth glass surface and cement paste [11, 13, 15]. Additionally, the smooth surface and minute water absorption of crushed waste glass particles resulted in a weaker cohesive force in the concrete mixture, which led to segregation and bleeding of the concrete [16, 17].

Additionally, applications to the concrete aggregates are partly limited and often not successful due to the problem of ASR, resulted in severely aggravating durability of concrete. The problem is fundamentally related to the increased solubility/instability of amorphous, disordered, poorly and micro-crystalline forms of silica in high pH [18, 19]. ASR is occurred, between a larger amount of alkali and reactive silica in the aggregates of waste glass as: (i) dissolved hydroxyl OH⁻ attaches to siloxane bonds (Si-O-Si) present at the silica surface, forming silanol (Si-OH) groups, (ii) these silica(te) reacts with alkalis (Na⁺ or K⁺) to form ASR gel, leading to expansion and ultimately causing concrete cracking [18-21]. The overall reaction is expressed as [20, 22]:

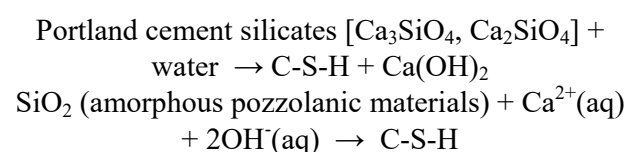


This reaction can occur with any aggregates that have reactive (amorphous) siliceous components. The presence of sodium and potassium alkalis is the primary chemical contributors to ASR [19, 20]. Hence, it is easily predicted that the increase in alkali content in concrete after the addition of waste mixed glasses as aggregate causes an ASR which is not beneficial to the performance of concrete, which provides the limited utilization of glass aggregate in concrete [23, 24]. Also, there are more microcracks and voids inside coarser waste glass compared to finer ones, and ASR expansion often occurs inside intrinsic microcracks and voids within waste glass particles, resulting from the increase in the risk of interaction between

silica and alkalis due to an increase in active specific surface of the aggregate [25]. However, the water absorption capacity of waste glass is negligible, so that the available moisture for evaporation in hydrated cementitious paste within the waste glass concrete core is lower than that in OPC [26, 27]. Consequently, low drying shrinkage occurs in waste glass concrete as well as the creep of concrete with waste glass aggregate is generally lower than that of OPC at long age [26].

Because of the risk of ASR as aggregates of waste glass on the concrete durability, considering waste glass contains large quantities of silica with an amorphous structure, another possible method of reusing waste glass has been also developed on the utilization as SCM including the economic and environmental concerns of waste glass utilization [28-33], if it is ground small enough [9] such as fly ash or ground granulated blast furnace slag.

The amorphous silica, in itself, possesses little or no cementitious property, but that will, in finely ground form and in the presence of moisture and calcium hydroxide [Ca(OH)₂] which is produced from the hydration of cement silicates [Ca₃SiO₄, Ca₂SiO₄], chemically react with Ca(OH)₂ at ordinary temperature to form secondary compound of calcium silicate hydrate [Ca₃Si₂(OH)₈] (C-S-H), which further improves the mechanical properties and durability of the hydrated cement paste [16, 34]. This is called pozzolanic reaction. The pozzolanic reaction of SCMs can improve concrete microstructure through changes in C-S-H composition and changes in the porosity [35]. The overall pozzolanic reaction scheme is as:



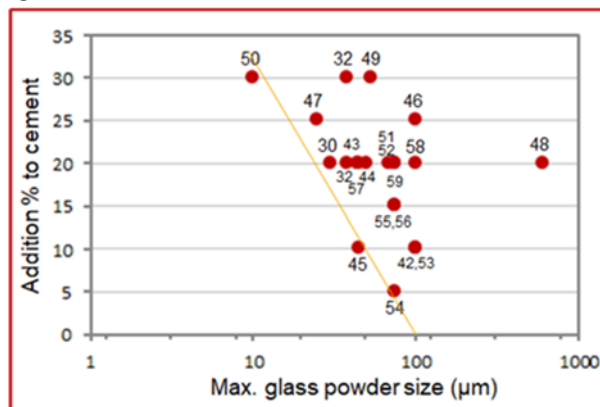
It is proved that when the waste glass is ground to a certain extent, it does not cause ASR, and it also has pozzolanic activity [28-30], and furthermore that glass powder has good pozzolan activity when the particle size is less than 20 μm [30] as cement, which can be improve the performance of concrete. When glass powder size is smaller, the pozzolanic reaction occurs between glass particle and Ca(OH)₂ instead of deleterious ASR [29]. Because fine glass powders have more reactive sites, these are less susceptible to ASR as a result of more complete pozzolanic reaction [36].

The particle size range in which glass begins to reduce ASR is ambiguous, however, a general average particle size in which reduction in ASR can begin to be seen below 1 mm [31, 37]. It is commonly considered that finely ground glass favors a relatively rapid pozzolanic reaction over the slower ASR [31].

The reason to suppress the ASR expansion by the pozzolanic reaction can be generally reviewed as: (i) Pozzolanic reaction between SCMs and cement hydrates contributes the reduced permeability of cement paste, and consequently the mobility of ions in concrete is reduced [20, 21, 38], (ii) The improved strength developed by the SCMs provides higher resistance to the expansive stress produced by ASR [21, 38], (iii) The secondary hydrates, C-S-H, produced by pozzolanic reaction, have the capacity to absorb and to entrap a significantly higher quantity of alkali ions than normal C-S-H, thus reducing the quantity of alkali ions and the pH in the pore solution [20, 21, 39, 40].

The pozzolanic activity of SCMs is related to their alkali binding capacity. Alkali binding, which occurs as a consequence of pozzolanic reactions, is accepted as the main mechanism of ASR mitigation by SCMs [39-41].

Table 2 and Figure show many results of previous researchers as SCM of grounded waste glass.



Relation between maximum glass powder size and partial replacement % to cement in previous results of researchers.

From the results shown in Fig. 1, variation of partial replacement % can be according to the size of waste glass powder, which may also results from the variation of chemical composition of waste glass used in the experiments of researchers. When the particle size distributions of the waste glass powder and the used OPC are almost same, the results of concrete strength with 20% replacement of cement are

Table 2

A summary of previous results for the use of waste glass powder as partial replacement to cement by researchers (R: residue, x_0 : mean diameter)

% waste glass	Glass powder, μm	Optimum %	Optimum glass powder, μm	Reference
30	< 38, 38-75, 75-150	30	< 38	32
20	< 20, < 40, 80-40, 100-80	20	< 20	30
0-20	1-100	10	1-100	42
20	44R=3% ($x_0=13$)	20	44R=3% ($x_0=13$)	43
10, 20	< 50	20	< 50	44
5,10,15	45R=10%	10	45R=10%	45
25	< 100 ($x_0=10-20, 40$)	25	< 100 ($x_0=10-20$)	46
25	< 25, 25-38, 63-75	25	< 25	47
5-20	< 600	20	< 600	48
10,20,30	38-900	30	< 53	49
20, 30	10R=12% ($800 \text{ m}^2/\text{kg}$)	30	10R=12%	50
20	69R=10% ($437 \text{ m}^2/\text{kg}$)	20	69R=10%	51
20	0-13, 13-38, 38-75	20	< 75	52
10,20,30	< 100 ($1169 \text{ m}^2/\text{kg}$)	10	< 100 ($1169 \text{ m}^2/\text{kg}$)	53
0-30	≤ 75 ($251 \text{ m}^2/\text{kg}$)	5	≤ 75	54
0-25	≤ 75	≤ 15	≤ 75	55,56
0-30	$\leq 38, \leq 45, \leq 75$	30, 20	$\leq 38, \leq 45$	57
0-60	100R=10% ($x_0=10$)	≤ 30	100R=10% ($x_0=10$)	58
0-25	74R=0.9%	≤ 20	74R=0.9%	59

also almost same compared to the concrete without waste glass [44]. Therefore, if waste glass is ground small enough such as fly ash or ground granulated blast furnace slag (size between 30 to 100 μm), with utilization of pozzolanic activity of waste glass powder, replacement to cement is certainly possible as SCM.

Based on the above results of previous researchers, the following regression equation can be derived from the lowest value of upper powder size at a certain replacement % to cement with a correlation coefficient of 0.91 as:

$$(\text{Replacement, \%}) = -30 \cdot \log_{10}(x_{\text{max}}) + 63$$

where x_{max} : upper limit size of glass powder (μm)

The optimum level of replacement of waste glass as SCM is actually obtained from the co-grinding of 80% (clinker: gypsum = 95:5) and 20% waste glass producing blended cement with about 400 m^2/kg of Blaine value, providing comparable mechanical properties of hardened cement paste [61]. Traditional SCMs such as fly ash are subject to wide variations of material composition according to the origin of coal mine. However, waste glass has a minor variation in chemical composition, and also is highly consistent which makes it a material of choice for recycling as SCM [43, 60]. Even though limited research has been conducted on

the durability of concrete with waste glass powder as SCM, the high pozzolanic reactivity of fine waste glass powder results in high concrete durability against water, chloride, and sulfate penetration [26, 27]. Concrete's resistance to acid attack and carbonation is also improved [26].

Conclusions and Perspectives

Applications to the concrete aggregates are partly limited and often not successful due to the problem of alkali-silica reaction (ASR), resulted in severely aggravating durability of concrete. Finely ground glass as oppose to coarse waste glass provides the pozzolanic activity with water and $\text{Ca}(\text{OH})_2$ obtained from the cement hydration, suppressing alkali-silica reaction. Besides improving the properties of concrete by pozzolan reaction by partial replacing, the waste glass powder will contribute to a greener environment in cement and concrete industries leading to global warming issues.

Guidelines on the comparative and relative descriptions of the fineness, adding ratio, strength, durability properties need to establish in glass powder blended cement and/or additional SCM of concrete for the recycling of waste glass as well as the reduction of CO_2 emission.

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