applied sciences

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Feasibility of Non-Remanufactured Waste Bottle Glass as Supplementary Cementitious Material

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Abstract: Theoretically, glass can be recycled entirely, but there are several requirements for remanufacturing. For the first time, this work studied industrial bottle waste glass (WG), which cannot be used to remanufacture new glass as a cement replacement for concrete applications. Wet and dry milling treatments were performed to reduce the particle size of WG and remove fibre and plastic contaminants. The different waste glasses treated were characterised by chemical, physical, and morphological analysis. Afterwards, mortar-level studies followed, using raw WG and wet-milled WG (AGWG) as a 10% Portland cement replacement. Mechanical and several durability indicators were assessed. WG and AGWG incorporation improved mortar performance against water capillary absorption, chloride ingress, and alkali–silica reaction. The unfavourable effect, namely, mechanical strength loss on glass-modified mortars, was mainly due to fibre contaminants observed by SEM on WG and AGWG samples. Even though wet milling reduced the amount and length of the fibre contaminants, it still did not guarantee adequate mechanical strength for the mortar. Thus, additional or alternative treatments, such as thermal treatments, must be explored.

Keywords: industrial waste glass; waste recycling; waste treatment; supplementary cementitious materials; cement; concrete

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Citation: Matos, A.M.; Sousa-Coutinho, J. Feasibility of Non-Remanufactured Waste Bottle Glass as Supplementary Cementitious Material. *Appl. Sci.* **2024**, *14*, 2004. https://doi.org/10.3390/app14052004

Academic Editor: Cesare Oliviero Rossi

Received: 8 January 2024
Revised: 12 February 2024
Accepted: 21 February 2024
Published: 28 February 2024

1. Introduction

Concrete, a mix of cement, aggregate and water, is essential to the basic needs of 21st-century society, such as housing, shelter, workplace, and transport infrastructure. Concrete is, in fact, the most consumed human-made material. This is because of the abundant raw constituent materials worldwide, the ability to set in various environments, and long-lasting durability and versatility [1]. The global demands for cement and concrete have been growing, mainly in emerging regions, motivated by rapid population growth and economic development [2].

Portland cement (PC) clinker is the most widely used binder for concrete production. The last forecast of the International Energy Agency Cement Roadmap estimates that cement demand may grow up to 23% by 2050 [3], and the Organisation for Economic Co-operation and Development admits that its demand might double by 2060 [4]. Unfortunately, the use of PC has a high environmental cost due to being responsible for the use of 7 billion tons/year of non-renewable natural materials and 2% and 5% of global primary and industrial energy consumption, respectively, resulting in 842 kg of CO_2 emissions, on average, per ton of Portland cement produced [5]. Worldwide, efforts have been made to reduce the environmental footprint of PC. The United Nations Environment Program Sustainable Building and Climate Initiative admits that the use of low-CO_2 supplements (supplementary cementitious materials—SCMs) as partial PC replacements and the optimisation of PC dosage in mortars and concrete products are the main pillars (considering a product-based approach) to substantially reduce cement and concrete global carbonic emissions over the next 30 years [5].

The partial replacement of Portland cement clinker with other materials, known as SCMs, has been under scrutiny [5–9]. SCMs include fly ash, clays, ground granular blast furnace slag, limestone filler, silica fume, and metakaolin [9,10]. Besides ecological and economic benefits, partial cement replacement by SCMs also offers engineering advantages, such as improving workability and segregation risk and reducing shrinkage and cracking risk. It may provide better mechanical strength and durability [9,11]. Limestone filler (LF), fly ash (FA), and granulated blast furnace slag (GBFS) presently constitute the overwhelming majority of SCMs, but the ready availability differs by country/region. Other SCMs, such as silica fume or metakaolin, are also used in cement/concrete but may not be readily available and are more expensive. For instance, in Portugal, LF is the available SCM on the market. Thus, new sources of SCMs are sought for further CO_2 reduction for clinker Portland substitutes [5].

In particular, it is essential to study waste or by-product materials that can partially replace clinker Portland (low-embodied CO_2 materials) [5–7], similar to what has been done in the past with other standardised SCMs [1]. An amorphous material constituted by silica, alumina, and/or lime can potentially become an SCM. So, beyond LF, GBFS, and FA, other materials, including industrial or agricultural wastes, can partially replace PC. Furthermore, using waste materials as SCMs diverts waste from landfills and gives rise to a value-added product, which offers additional economic and ecological benefits [5,12,13]. The main barriers are the lower reactivity of these alternative materials compared with cement [14] and the need for additional chemical/physical treatments to increase their reactivity [15]. Moreover, waste materials or by-products may present contaminants or elevated alkali levels, which may provide deleterious reactions when incorporated into cement-based materials, which limits the actual use of such materials [15].

Glass is a versatile material and part of society's daily life in many fields and for different purposes, such as packaging or container and decoration (e.g., bottles, jars), flat glass (e.g., windows, windscreens), bulb glass (e.g., light globes), and cathode ray tube glass (e.g., TV screens, monitors) [16]. Glass products or elements often have a limited useful life and are discharged [17–19]. Theoretically, glass can be recycled entirely, but there are several requirements for remanufacturing. Thus, the glass fraction that cannot be introduced in the remanufacturing process is usually disposed of in landfills [16]. About 200 million tons of glass are estimated to be landfilled worldwide annually, with 80% of the glass disposal being soda–lime glass, widely used in beverage bottles [16]. But glass is a non-biodegradable material; thus, waste glass can take one million years to break down naturally when landfilled [16]. Therefore, recycling waste glass has been a global concern to the scientific community [20].

Recycling glass waste in construction material, particularly crushed glass, has been under scrutiny over the last 70 years. Various applications can be found in a literature survey, such as using crushed waste glass as aggregate in subbase and unbound base mixtures, aggregate in asphalt mixtures, and SCMs and/or aggregate in cementitious materials. Ground waste glass mainly consists of silicon and calcium with an amorphous structure. Previous research shows that glass powder can be considered a pozzolanic–cementitious material according to the chemical requirements in ASTM C 618 and NP 4220; only the alkali content fails [21,22]. The pozzolanic properties of ground waste glass, when very fine grounded, motivated several researchers to use it as a partial cement replacement [16,20–25]. Therefore, the main findings concerning recycling waste glass powder (finely milled waste glass, glass powder—GP) as a cement replacement for concrete applications in terms of mechanical and durability performance are described below. More detailed information is available in exhaustive review articles [15,17,20].

Incorporating glass powder (GP) generally improves the workability of fresh mortar or concrete [21–23,26–28]. Other studies argue that waste glass influences cement replacement on slump and other engineering properties depending on the glass powder type and properties [29,30]. Idir et al. [31] studied the compressive strength development of mortars with glass powder as a cement replacement for up to 90 days, varying glass powder fineness and incorporation dosage. The results revealed that increasing GP dosage from 10% to 40% reduced compressive strength gradually. However, the pozzolanic reactivity of GP was observed after 28 days since the pozzolanic reaction is enhanced with particle size reduction, resulting in significant strength increments at later ages. According to the literature, compressive strength generally decreases slightly with the increase in GP dosage, and the optimum content seems to be between 10–30% of GP used as a cement replacement. However, the finer the glass used, the higher the concrete strength, especially at a late age (90 days) [15,20,21,23–25,32]. In fact, past research evidence that glass powder pozzolanic reactivity depends on particle size. Generally, glass powder particles below 75 µm were very reactive, especially below 38 µm [16,31,33–39]. The pozzolanic reactivity of GP is governed by the particle size [21]. Glass powder particle size is also a key factor regarding deleterious alkali–silica reaction (ASR) expansion. The body of literature states that particles below 300 microns do not contribute to the reaction [17,40,41]. In fact, micro-sized glass particles can even mitigate expansion due to the ASR [22,25,42].

Previous research is also unanimous concerning improved durability when glass powder is used as a cement replacement, namely rising resistance against chloride ion penetration [21,22,24]. Increased GP content showed higher resistance to chloride ion penetration in mortar specimens [28,34,43]. Pore refinement by pore blocking and pore filling resulted in less permeable concrete containing GP, thus locking chloride ion penetration [44]. Compared with fly ash, GP improved chloride resistance but was less efficient than silica fume [21,22,24,29]. Higher carbonation depth on concrete/mortars were reported with the incorporation of GP [21,22,24] owing to the increased porosity at early ages and lower amount of $Ca(OH)_2$ in the cementitious matrix, which is typically observed when SCMs are used [30].

The increased GP dosage of up to 20% as a cement replacement in mortar reduced water absorption, which is attributed to the higher density of GP mortar mixes [30,36]. The effect of GP on pore refinement and pozzolanic reactivity was well pronounced based on the depreciated water absorption values and voids ratio in GP-modified concrete [35]. A literature survey showed similar sorptivity results on reference and GP mortars, with 10% and 20% GP as cement replacements [21]. Other studies observed lower sorption in concrete incorporating GP due to the dense microstructure resulting from CSH formation and increased pozzolanic reaction [28,45]. In addition, water penetration depth was lower in concrete with up to 60% glass powder than in the control mix [26,34].

Research Significance and Objectives

The use of glass in concrete production in large quantities needs an adequate characterisation of waste glass properties (chemical and physical) and an understanding of the waste glass effect on final concrete product engineering properties [15,20,46].

Based on the literature survey, cement-based materials incorporating ground waste glass seem promising. However, research is still required to set a guideline for the appropriate use of ground waste glass in cement-based materials. Several glass types may present dissimilar chemical compositions, originating different effects on the final cement-based material product. Therefore, it may be necessary to establish the pre-treatment process before application.

Theoretically, glass can be recycled entirely, but there are several requirements for remanufacturing. For the first time, this work studied industrial bottle waste glass (WG), which cannot be used to remanufacture new glass as a cement replacement for concrete applications. The first stage of this study consisted of a deep analysis of WG and laboratory treatments to remove (still present) fibre and paper contaminants and improve pozzolanic properties. The second phase studied the mechanical and durability properties of treated wastes in binary blended cement mortars using waste glass as a cement replacement.

2. Methodology

2.1. Non-Remanufactured Waste Bottle Glass

2.1.1. Origin

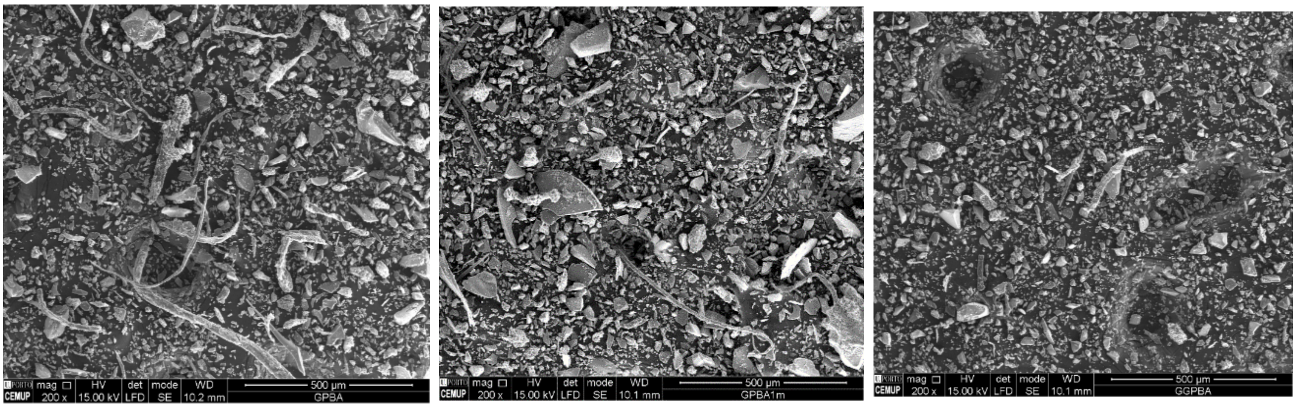
The industrial waste glass used in this work is waste produced by BA Glass Portugal, S.A. (Avintes, Portugal), resulting from the company's glass bottle recycling process. In brief, the bottle's cullet is dried, and the ferrous and non-ferrous contaminants (e.g., metals, stones, ceramics, porcelains, opals, organic waste, plastics, and other glass) are collected and then milled in different sizes. The finer glass waste was used in this study.

2.1.2. Laboratory Waste Glass Milling

Waste glass raw samples were received in the laboratory and dried until a constant mass. Then, the samples were sieved with a 1 mm sieve to separate the waste glass powder from small visible amounts of paper, plastic, and fibres, as shown in Figure 1. The materials passed through a 1 mm sieve were used in the current research and named waste glass (WG), as shown in Figure 2. The scanning electron microscopy (SEM) images of WG obtained using a secondary electron mode are presented in Figure 3a.

Figure 1. WG contaminants retained in the laboratory via a 1 mm sieve.

Figure 2. WG passed through a 1 mm sieve.



(**a**) (**b**) (**c**)

Figure 3. SEM (secondary electron mode) images of glass powders: (**a**) WG; (**b**) GWG; and (**c**) AGWG.

The use of waste glass as a cement replacement is better as the particles are finer, than cement. This is because the finer the powder, higher the pozzolanic activity. Thus, the WG was ground in the planetary mill RETSCH PM100 in the following steps: (a) grinding using 20 mm diameter zircon balls for 15 min at 350 rpm and (b) grinding the resulting material from the previous step using 1 mm diameter zircon balls for 15 min at 350 rpm speed. This material was named ground waste glass (GWG); the samples were then taken for physical analysis. The particle size analysis and the specific surface results (Blaine method), see Table 1, revealed that the ground material (GWG) was coarser than the original WG. It was concluded that fibres (see Figure 4) created tangled lumps during milling, compromising both the particle size analysis method and the Blaine test (Table 1). SEM observation confirmed the existence of fibres, as depicted in Figure 3b.

Table 1. Density, specific surface, and particle size distribution of cement, WG, GWG, and AGWG.

Materials	Cement	WG	GWG	AGWG
Density (kg/m^3)	3110	2280	2280	2280
Specific surface (cm^2/g)	3742	3462	3131	4779
d_{10} (µm)	1.42	4.45	4.57	2,92
d_{50} (µm)	13.50	25.96	32.96	18.34
d_{90} (µm)	40.52	127.00	197.00	82.83

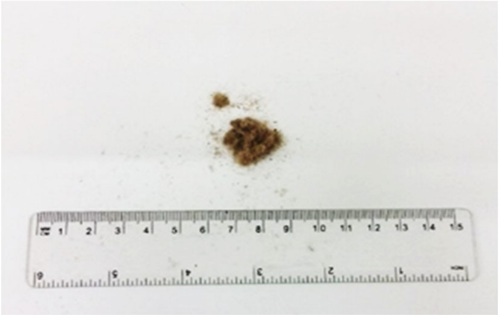


Figure 4. Fibre contaminants found in dry-milled ground WG (GWG).

Thus, it was decided to repeat the second step of the milling process using alcohol. Even though it is a more time-consuming process due to the drying needed after milling, previous research by authors obtained good results using this method. After 15 min of grinding with alcohol at 350 rpm, the sample was dried in an oven at 50 °C for 10 min and then mechanically sieved to remove the spheres. The SEM observation of the resulting material, alcohol-ground waste glass (AGWG), revealed that even after grinding with alcohol, the waste glass contained fibres. However, the fibres seemed to be shorter, see Figure 3c.

2.1.3. Laboratory Waste Glass Incineration

Given the results obtained in milled waste glass presented and discussed in Section 3, namely the mechanical strength and SEM observations, it was concluded that fibre contaminants caused a significant impact on cementitious mortar properties. It seemed that dry or wet milling might not be enough. Thus, it was considered to incinerate the WG material at a temperature of 700 °C for 6 h. Afterwards, the incinerated material was homogenised in the planetary mill (as described in Section 2.1.2). Mortar specimens 40 × 40 × 160 mm^3 were produced to assess the mechanical strength according to EN 196-1 at 7 and 28 days on a mortar with 10% cement replacement of incinerated WG (WG700).

2.1.4. Waste Glass Properties

As refered, Table 1 summarises the physical properties of cement WG, GWG, and AGWG: density (determined by E-64), specific surface (determined according to EN 196-6, Blaine method), and particle size distribution (determined via laser method). The major elements analysis of cement and WG is presented in Table 2 and was determined by FRX.

Table 2. Main oxide composition of cement and WG.

Materials	**Chemical Composition (% by Mass)**												
	SiO_2	**CaO**	**Fe_2O_3**	**Na_2O**	**Al_2O_3**	**MgO**	**K_2O**	**TiO_2**	**P_2O_5**	**Cl**	**SO_3**	**Insoluble Residue**	**LOI**
Cement	20.33	63.31	3.02	0.20	4.83	1.91	0.73			0.05	2.85	1.03	2.28
WG	60.91	10.78	1.15	9.63	2.54	1.07	0.92	0.15	0.16	0.16	0.49		12.27

As can be perceived from Table 2, WG is mainly composed of silicium oxide (61%), calcium oxide (11%), and sodium oxide (10%). Based on the literature survey, the percentages of the main constituents of glass do not differ widely, except Al_2O_3 and CaO. Soda–lime glass comprises approximately 70% silica, 13–17% Na_2O, and 10% CaO [16,21–23]. The WG used in the current work presented less silica and higher calcium content. This might be related to the contaminants still present in WG, giving rise to higher calcium content and LOI (12%, see Table 2) due to organic matter.

The use of waste glass powder as an SCM for cement-based materials/concrete does not yet have a dedicated standard. As such, the following standards were considered as a benchmark: (a) a general Portuguese standard for pozzolanic materials, NP 4220, and (b) an American standard, ASTM C 618, regarding fly ash and raw or calcined natural pozzolan for cement. The requirements of those standards are explained in Table 3. Bold and/or underlined cells in Table 3 highlight requirements WG did not comply with. Even though WG used in the current research presents a composition similar to natural pozzolan and other waste glasses, the requirements were not met. ASTM C 618 (class N and F) stipulate a minimum content of 70% for the sum of SiO_2 + Al_2O_3 + Fe_2O_3. Although not far, WG did not reach 70% of SiO2 + Al_2O_3 + Fe_2O_3.

According to ASTM C 618, the LOI presents a maximum limit of 10% for class N and 6% for classes F and C. Similarly, NP 4220 specifies a maximum LOI of 10%. The LOI of WG did not comply with any of those standards.

Concerning the sulphate content, an average SO_3 content of 0.48% was obtained for the WG sample, which is very low compared with the benchmarking limits referred by NP 4220 (3.0%) and ASTM C 618 (4.0% minimum limit).

A maximum chloride content of 0.1% is imposed by NP 4220. The WG was found to have an average chloride of nearly 0.1% (see Table 2). Since waste glass powder will probably be used as partial cement replacement in a percentage lower than 10–20%, chloride content might not be a concern.

Regarding the physical properties presented in Table 1, the PSDs of WG and AGWG are higher than cement. The d_{90} values were 40, 127, and 82 μm for cement, WG, and AGWG, respectively, and the d_{50} values were 13, 26, and 18 μm. A lower PSD would benefit pozzolanic ability [16,31–38] since past research showed that the smaller the particle size, the higher the pozzolanic reactivity of the glass powder particles [39].

Table 3. Chemical requirements for pozzolanic SCMs for concrete, according to NP 4220 and ASTM C 618 (in bold and underlined cells, requirements failed for WG on both standards; in bold cells, requirements failed for WG on one of the standards).

	WG	NP 4220	ASTM C 618		
		Pozzolans in General	Fly Ash and Pozzolan		
			N	F	C
$SiO_2 + Al_2O_3 + Fe_2O_3$ (%)	**64.60**	-	>70	>70	>50
CaO (%)	**10.78**	<10.0	-	-	-
Cl (%)	**0.16**	<0.1	-	-	-
SO_3 (%)	0.49	<3.0	<4.0	<5.0	<5.0
$(Na_2O)_{eq}$ (%) $(Na_2O)eq(\%) = Na_2O + 0.658K_2O$		<5.0	-	-	-
Loss on ignition (LOI) (%)	**12.27**	≤5.0	<10	<6.0	<6.0

2.2. Mortar Study

2.2.1. Mortar Specimen Production

The waste laboratory treatment was followed by a study at the mortar level, which was conducted using different waste-treated glasses as a cement replacement. The following raw materials were employed for the mortar study: Portland cement CEM I 42.5R, under the EN 197-1, consisting of a percentage equal to or greater than 95% of clinker with the remaining percentage consisting of additional minor cement constituents; WG and AGWG (described in Section 2.1); CEN sand, following the EN 196-1, which is natural, siliceous sand with rounded grains, with silica content equal to or greater than 98%; and distilled water was used as prescribed in EN 196-1 at room temperature.

Three mortar types were studied according to mixture compositions presented in Table 4. The nominated mortars, WG10, AGWG10 and WG700, refer to binary blend mortars, including WG, AGWG and WG700, as 10% partial cement replacement (by mass), respectively (see Table 4). A reference mortar was also produced, including only cement as a binder (REF in Table 4). The mortar mixing procedure, casting, and compaction followed the steps described in EN 196–1. The specimens remained for 24 h in the stainless steel moulds and were then maintained in water curing in a fog room (temperature 20 ± 2 °C and HR > 95%) until testing. The ASR mortar bar test (ASTM C 1567) required a specific curing process, further explained in Section 2.2.2.

Table 4. Mortar mixture proportions under study.

Mortar ID	Cement (g)	WG (g)	AGWG (g)	WG700	Water (g)	Sand (g)
REF	450	-	-	-	225	1350
WG10	405	45	-	-	225	1350
AGWG10	405	-	45	-	225	1350
WG700	405	-	-	45	225	1350

2.2.2. Tests on Mortars

Mechanical strength from 7 to 84 old days and the following durability indicators were assessed on the hardened mortars REF, WG10, and AGWG10: electrical resistivity, water absorption by capillarity, resistance to chloride migration, carbonation resistance, and risk of alkali–silica reaction. Table 5 summarises the experimental testing plan, and the test methods employed are described as follows.

Table 5. Experimental testing plan summary.

	Test Standard	Curing Regime	Testing Age	Number of Samples for Each Age and Each Mortar Type	Sample Geometry and Size
Mechanical strength	NP EN 196-1	Water curing at 20 °C	7, 28, 56, and 84 days	3	Prismatic $40 \times 40 \times 160$ mm^3
Electrical resistivity	Two-electrode method		7, 28, 56, and 84 days	3	Prismatic $40 \times 40 \times 160$ mm^3
Water absorption by capillarity	RILEM TC 116-PCD		42 days (28 days wet curing + 14 days at 40 °C, until constant mass achieved)	3	Prismatic $40 \times 40 \times 45$ mm^3
Resistance to chloride migration	NT Build 492		28 days	3	Cylindrical h = 50 mm, Ø = 100 mm
Carbonation resistance	RILEM CPC-18		63 days (14 wet curing + 14 days at 20 °C and HR 60% + 35 days CO_2 exposure)	3	Prismatic $40 \times 40 \times 160$ mm^3
Risk of alkali–silica reaction	ASTM C 1567 adapted	1-day water curing at 80 °C (according to ASTM C 1567)	Up to 28 days	3	Prismatic $25 \times 25 \times 250$ mm^3

Mechanical Strength

Three prismatic specimens ($40 \times 40 \times 160$ mm^3) were produced for each mortar type (see Table 4) and age, namely, 7, 28, 56, and 84 days, and tested according to EN 196-1. Specimens were kept under water curing (20 °C) until the testing date.

Electrical Resistivity

The electrical resistivity for mortars (presented in Table 4) between 7 and 84 days was measured on $40 \times 40 \times 160$ mm^3 specimens for each mortar type presented in Table 4. The specimens were kept under water at a controlled temperature (20 ± 2 °C) and removed to perform the resistivity test at established ages.

At two opposite surfaces of the specimen, stainless steel grids were embedded (during casting) and used as electrodes (see Figure 5). For the two-electrode resistivity method assessment, alternating current was preferred. As such, a signal generator performed a sinusoidal wave with a peak voltage of ±10 V and a frequency of 100 Hz. Then, a digital multimeter (Keithley) measured the resulting current. The electric resistivity (ρ), Ω m , was calculated by multiplying the resistance by the cell constant, which for prismatic specimens is the ratio of the surface area to the length. The following equation applies:

$$\rho = \frac{VA}{IL} \tag{1}$$

where V, voltage (volts); I, current (A); L, length (m); and A (m^2) is the cross area of the test specimen through which current passes. The resistivity results were considered the average value from three specimens tested at each age.

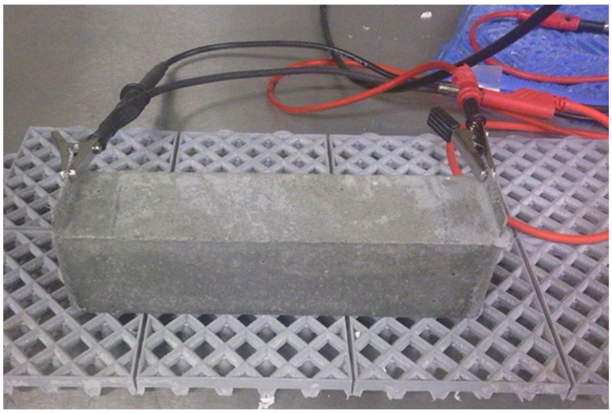


Figure 5. Mortar resistivity measurement.

Water Absorption by Capillarity

The RILEM recommendation TC 116-PCD was adopted to measure the water absorption by capillarity on three replicate prismatic specimens for each mortar type (Table 4). Firstly, the specimens were dried at 40 °C until constant mass. For the test, dried specimens were placed in a water bath with automatically adjusted water level at a constant depth of approximately 3 mm (see Figure 6). Each sample mass was measured before partial water immersion (0 min) and at the following times after contact with water, 5, 10, 20, 30, 60, 90, 120, 180, and 240 min.

Figure 6. Water absorption by capillarity conducted in a water bath with an automatically adjusted water level.

The following equation determines the sorptivity, S, of each mortar specimen during testing time:

$$A = A_0 + S \times t^{0.5} \tag{2}$$

where A_0 (mg/mm^2) is the water absorbed initially by pores in contact with water, A (mg/mm^2) is the water absorption by unit area of the mortar specimen's surface from the moment the specimen is dipped in water, and t is the elapsed time from the moment the specimen makes contact with water (in minutes).

Rapid Chloride Migration Test

The NT Build 492 procedure was followed to access the chloride ion penetration resistance. The test was performed at 28 days on three replicate cylindrical specimens for each mortar under study (Table 4). Before testing, the samples were vacuum-soaked with a $Ca(OH)_2$ saturated solution. Afterwards, an electrical potential was applied across each specimen to force the chloride ions in the catholyte solution (10% NaCl) to migrate through them, as shown in Figure 7. In this case, a potential of 25 V was applied, and the test was performed for 24 h. Afterwards, the cylindrical samples were axially split into two halves, and silver nitrate solution (0.1 M) was sprayed on one of the surfaces. This solution precipitated the chloride salt into white silver, as seen in Figure 8.

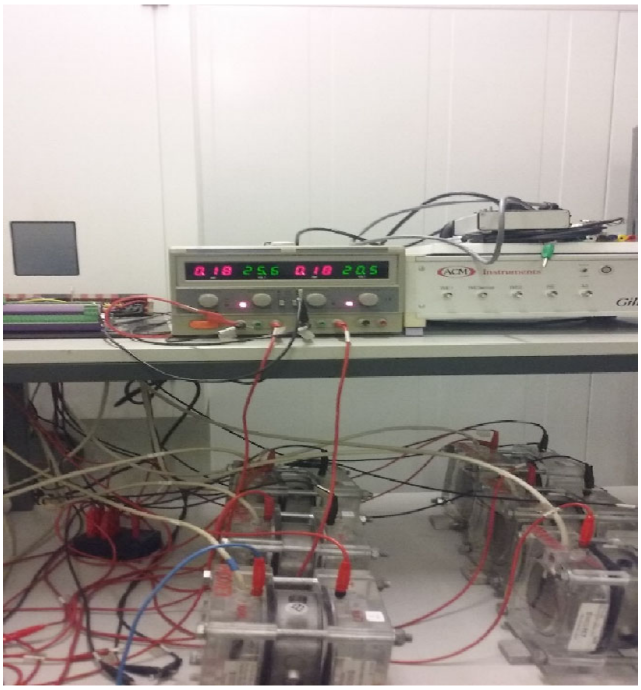


Figure 7. Chloride migration test set-up.

Figure 8. White silver precipitation of chloride salt.

Equation (2) allows the calculation of the non-steady-state migration coefficient, D_{nssm}:

$$D_{\text{nssm}} = \frac{0.0239(273+T)L}{(U-2)t} \times \left(x_{\text{d}} - 0.0238\sqrt{\frac{(273+T)Lx_{\text{d}}}{U-2}} \right) \times 10^{-12} \tag{3}$$

T is the mean value of the initial and final temperatures in the 0.3 M NaOH solution; U is the absolute value of the voltage applied (V); L is the sample thickness (mm); t is the duration of the test (min); and x_d is the mean value of the penetration depths (mm).

Accelerated Carbonation Test

The resistance to CO_2 was evaluated according to RILEM CPC-18. Three prismatic specimens ($40 \times 40 \times 160$ mm^3) were cast for each mortar type (see Table 4). The specimens were water-cured for 14 days and then dried in a controlled environment room

(T = 20 ± 2 °C and HR = 50 ± 3%) for 14 days. After that, the specimens were exposed to accelerated carbonation in a climatic chamber with a CO_2 concentration of 5 ± 0.1%, RH = 60 ± 5%, and temperature of 23 ± 2 °C. At the 35th day of exposure to CO_2, a 1 cm slice thickness was cut from each sample and sprayed with 1% phenolphthalein solution. Exposure to a carbonation environment reduces the pH of the cementitious system mainly due to the reaction of calcium hydroxide with CO_2. As a result, the phenolphthalein solution remains colourless in the carbonated region and red/pink in the non-carbonated region [44].

Expansion Due to ASR

The risk of alkali–silica reaction occurrence due to waste glass incorporation was monitored according to ASTM C 1567 with two slight modifications [21,22,25]. Instead of using the mortar proportioning specified in the standard, the specimens were produced using mortar mix proportions presented in Table 4. In addition, mortars were produced using CEN sand according to EN 196-1. In brief, the ASR linear expansion procedure is as follows: three prismatic specimens (25 × 25 × 285 mm^3, see Figure 9) were produced for each mortar type (Table 4) and demoulded from steel moulds after 24 h. Afterwards, specimens were submitted to accelerated curing in a water bath at 80 °C for 24 h. At that time, specimen lengths were assessed using a digital indicator with an accuracy of 0.001 mm. This length was considered the initial length of each specimen (L_0). Then, the mortar specimens were introduced into a NaOH solution and linear length was measured at pre-defined ages up to 28 days.

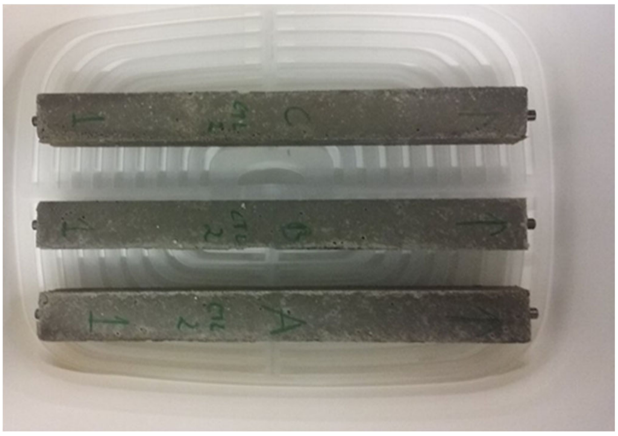
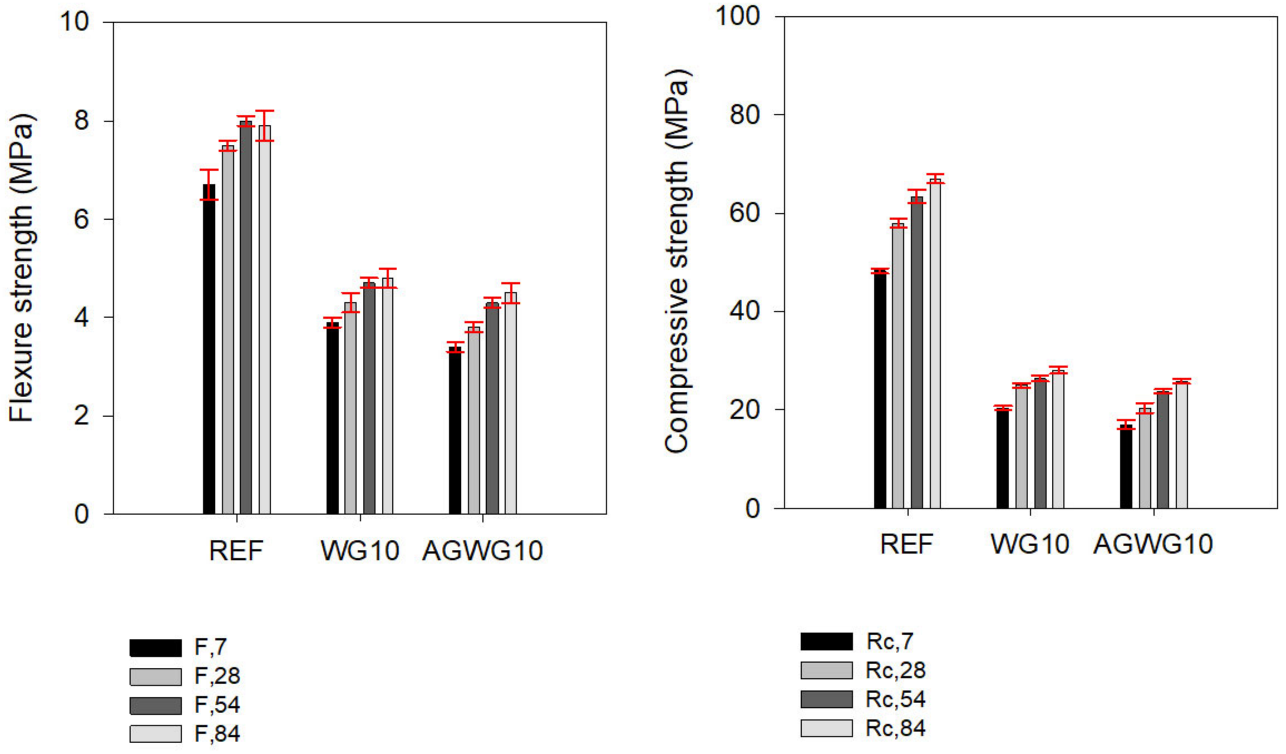


Figure 9. Mortar specimens used in the ASR test.

3. Results and Discussion

3.1. Mechanical Strength and Strength Activity Index

Figure 10a,b depict the flexural and compressive strength of mortar specimens at 7, 28, 54, and 84 days, respectively. As can be perceived, the mechanical strength of the mortar specimens, including glass powder, WG10, and AGWG10, significantly decreased. Compressive strength of 58.3, 20.4, and 25.0 Mpa were obtained for REF, WG10, and AGWG10 mortars at 28 days, respectively, while at 86 days, the results were 67.0, 28.1, and 25.8 Mpa, respectively. Additionally, Table 6 presents the strength activity index (SAI) at 7, 28, 54, and 86 days for the WG10 and AGWG10 mortars (bold and underline cells refers to SAI at 28 days). The activity index for the reference age of 28 days was about 43% and 35% for the WG10 and AGWG10 mortars, respectively. Considering benchmarking, ASTM C 618 states a minimum of 85% of SAI at 28 days, while NP 4220 prescribes an SAI that is equal, at least, to the cement percentage in the modified mortar. This means that, in this study, an SAI of 90% must be obtained since WG10 and AGWG10 contain a binder with 90% cement (and 10% cement replacement). As can be perceived, the mechanical performance of mortars with waste glass was far beyond the expected and did not corroborate with past research [15,20–22,24,25,41]. However, the waste glass powder employed in the current work has particularities compared with the literature survey. The chemical composition is slightly different, as discussed in Section 2.1.4, and some contaminants seem to affect the mortar's mechanical behaviour. The presence of fibre contaminants (as presented in Section 2.1.2 and Figure 3) reduced size during the wet milling process (see Figure 4), but significantly interfered with mechanical strength, compromising the use of these binary binders for structural applications.



(**a**) (**b**)

Figure 10. Mechanical strength at 2, 7, 54, and 86 days: (**a**) flexure; (**b**) compressive.

Table 6. SAI for the WG10 and AGWG10 mortars at 7, 28, 54, and 86 days.

Age (Days)	WG10	AGWG10
7	42.2%	35.2%
28	**43.1%**	**35.2%**
54	41.7%	37.5%
86	41.9%	38.5%

Figure 11 shows the mechanical strength results of incinerated waste glass-modified mortars (WG700). As can be perceived, the incinerated WG as a cement replacement resulted in a similar performance of the REF mortar both in flexure and compression. The compressive strength was 51.5 and 49.9 Mpa for the REF and WG700 mortars, respectively. This resulted in an SAI of 96.9% at 28 days, which indicated that the milling treatment (Section 2.1.2) was insufficient to obtain a glass powder without impurities (namely, fibres). However, incineration treatment of this particular waste may be an option. Considerations of cost and technical viability must be reflected and balanced.

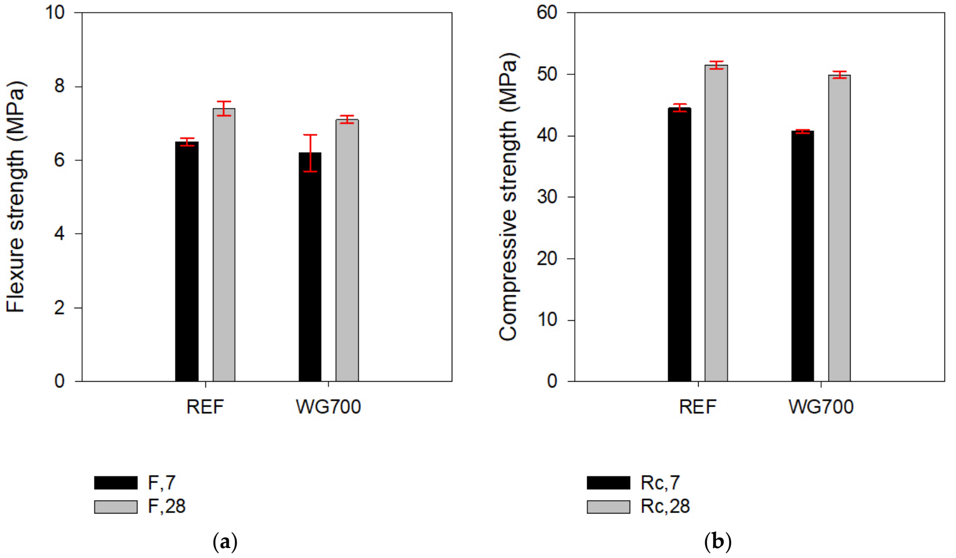


Figure 11. Mechanical strength at 7 and 28 days: (**a**) flexure; (**b**) compressive.

3.2. Durability Indicators

According to published studies, the durability of mortars/concretes with glass powder is usually improved. The durability-related results obtained in this work are described below.

Electrical resistivity measurement may be a simple indicator of the durability of cement-based materials [9,16]. The electrical resistivity depends on the pore structure, such as porosity and pore connectivity, and pore solution conductivity. As such, electrical resistivity provides valuable information regarding cementitious materials' microstructure and hydration process [29,47,48]. Figure 12 shows the electrical resistivity results of mortars between 7 and 86 days. At an early age, 7 days, the resistivity was similar among mortars under study. Afterwards, a substantial increase for glass powder mortars occurs, reaching, at 86 days, 65.2 and 62.9 ohm-metre for WG10 and AGWG10, respectively, while for the REF mortar, the result was 46.3-ohm metre. This translates as an increase of about 40% for glass powder mortars. The enhancement in the electrical resistivity of cement pastes modified with waste glass can be attributed to pore refinement in the microstructure resulting from the eventual pozzolanic behaviour of WG and AGWG. This corroborates past research [29,47,48]. Figure 12 also suggests that the resistivity of glass mortar mixtures will continue to increase beyond 84 days.

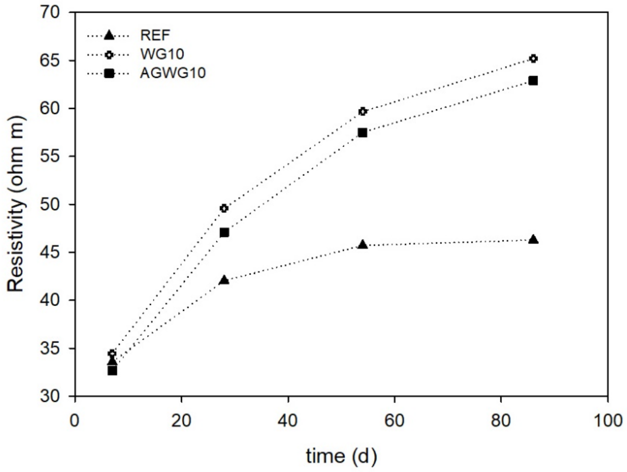


Figure 12. Electrical resistivity between 7 and 86 days.

The average sorptivity was 0.0490, 0.0370, and 0.0440 mg/(mm^2 × $min^{0.5}$) for the REF, WG10, and AGWG10 mortars, as presented in Table 7. Using waste glass powder slightly improved the capillary water absorption of mortars, namely WG. The pozzolanic reaction provided by the reaction of GP with calcium hydroxide seemed to decrease the porosity of mortars. Previous studies also observed decreased sorptivity with increased GP content [28,45].

Table 7. Sorptivity results for mortars under study.

Mortar ID	Sorptivity (mg/(mm^2 × $min^{0.5}$))
REF	0.0490 ± 0.003
WG10	0.0370 ± 0.003
AGWG10	0.0440 ± 0.008

The results from the rapid chloride migration test are presented in Table 8, including the chloride penetration depths (X_d) and the corresponding average values of non-steady-state chloride migration coefficients, D_{nssm}. As can be perceived, the D_{nssm} was similar among the mortars, being slightly higher for the WG10 mortar, followed by REF and ASGW10. As such, WG slightly decreases resistance to chloride ion penetration, which might be related to higher PSD than cement and AGWG (see Table 2), as discussed in Section 2.1.4. Moreover, these results were not expected. Improved performance on cement-based mortars against aqueous chloride solution is reported in the literature, as discussed in Section 1.

Table 8. Rapid chloride migration test results of obtained mortars under study.

Mortar ID	xd (mm)	U (V)	D_{nssm} (10^{-12} m^2/s)
REF	22.07	25.27	14.26
WG10	25.23	25.36	15.98
AGWG10	20.85	25.00	13.50

A decrease in carbonation resistance was noticed for mortars with the incorporation of glass powder, as presented in Table 9, which follows previous findings [21,24,25] and is typically observed on some SCMs when used as a cement replacement [44], which is probably due to CH reduction.

Table 9. Carbonation depth results obtained for mortars under study.

Mortar ID	Carbonation Depth (mm)
REF	3.58 ± 1.81
WG10	9.30 ± 1.65
AGWG10	11.30 ± 1.62

Figure 13 depicts the ASR linear length change of the REF, WG10, and AGWG mortars. According to ASTM C 1567, a mortar with an expansion lower than 0.10% after 16 days (and 14 days after immersion in 1 N sodium hydroxide solution at 80 °C) is expected to have a low risk of deleterious expansion. The mortar mixture is considered potentially expansive if the expansion exceeds 0.10 % at 16 days after casting. The expansion limit of 0.1% is highlighted in Figure 13 (horizontal dotted red line). When Portland cement is the binder (REF mortar), the mixture is classified as potentially expansive according to ASTM C 1567. Mortar prisms presented an expansion of 0.10% after 14 days of immersion in NaOH at 80 °C. Concerning WG10 and AGWG10, glass powder mitigates deleterious ASR expansion compared with the REF mixture, with expansion values of 0.05 and 0.03%, respectively, after 14 days of immersion in NaOH at 80 °C. Figure 13 also suggests that the expansion of the REF mortar will continue to increase beyond 28 days, while in glass-modified mortars it was stable for 16 days.

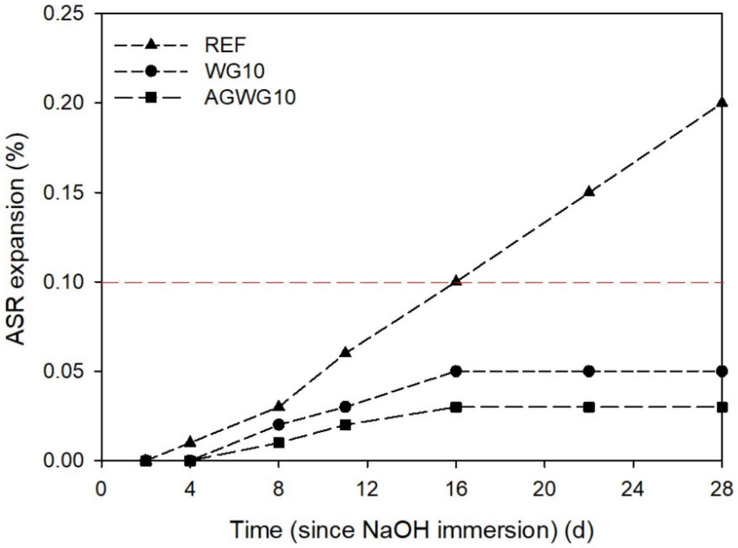


Figure 13. ASR expansion up to 28 ageing days.

These results are in accordance with previous findings in which micro-sized glass particles do not contribute to ASR expansion and can even mitigate it [17,22,25,40–42]. According to Taha and Nounu [49], the reactive silica available in glass powder reacts with $Ca(OH)_2$ very quickly and forms additional CSH mineral phases. Therefore, the reactive silica of glass powder will no longer be available for the ASR expansion, which occurs in later stages.

4. Conclusions and Final Remarks

The potential use of non-manufactured waste bottle glass as an SCM for concrete was investigated. Firstly, WG obtained from a local bottle industry was dried, sieved, and ground to obtain a PSD similar to cement. Since dry milling did not present reasonable results (GWG), wet milling followed (AGWG). Chemical and physical properties of WG and AGWG, as well as SEM observations, were carried out. In the second stage, mortar-level studies were performed, including WG and AGWG as a surrogate replacement, and mechanical behaviour and durability properties were evaluated. The main conclusions can be summarised as follows:

- The main oxide composition revealed that WG consists mainly of SiO_2, CaO, and Na_2O.
- A substantial decrease in mortar mechanical strength occurred when WG or AGWG were used as a 10% mass cement replacement.
- The mechanical strength of mortar, including 10% of incinerated waste glass powder, (WG700) achieved the REF mortar levels.
- Electrical resistivity was higher on glass-modified mortars, indicating the possibility of glass powder pozzolanic reactivity.
- The resistance against chlorides and capillary water absorption was similar among all mortars under study.
- ASR was reduced by over 30% in mortar specimens with 10% WG or AGWG as a cement replacement.

The presence of fibres due to labels in recycled bottles seems responsible for the lower mechanical strength of mortars WG10 and AGWG10. A global analysis of the tests also showed that the WG mortars generally presented higher durability than those with the wet-milled material (AGWG) and even the REF mortars (excluding the carbonation test), implying that grinding may not be a solution to be considered. Given these results, the possibility arose of burning the material to eliminate the fibres and perform mortar strength tests to replace the incinerated material. Thus, finding the most eco-friendly and cost-effective way to treat WG is essential. Therefore, thermal treatment of WG may need to be incorporated in PC-based composites to deplete fibres and other contaminants. This will probably lead to a more successful application of this waste as an SCM for conventional concrete mixtures.

Author Contributions: Validation, A.M.M. and J.S.-C.; Formal analysis, A.M.M.; Data curation, A.M.M.; Writing—original draft, A.M.M.; Writing—review & editing, J.S.-C.; Project administration, A.M.M. and J.S.-C.; Funding acquisition, A.M.M. and J.S.-C. All authors have read and agreed to the published version of the manuscript.

Funding: This work was financially supported as follows: Base Funding—UIDB/04708/2020 with DOI 10.54499/UIDB/04708/2020 (https://doi.org/10.54499/UIDB/04708/2020); Programmatic Funding—UIDP/04708/2020 with DOI 10.54499/UIDP/04708/2020 (https://doi.org/10.54499/UIDP/04708/2020) of the CONSTRUCT—Instituto de I&D em Estruturas e Construções; national funds through the FCT/MCTES (PIDDAC); and FCT—Fundação para a Ciência e a Tecnologia through the individual Scientific Employment Stimulus 2021.01765.CEECIND.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available in the article.

Acknowledgments: The collaboration and materials supplied by Secil and BA Glass Portugal are gratefully acknowledged. The authors especially thank Engineer Vasco Neves for laboratory work support.

Conflicts of Interest: The authors declare no conflict of interest.

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