Conversion of Hard-to-use Wastes to New Raw Materials for Low-Energy Glass/Mineral-Wool Manufacturing

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ABSTRACT

During the pyrometallurgical processes, a substantial quantity of high-temperature ironmaking and steelmaking slags, generated at temperatures ranging from 1400 to 1600 ℃, is wasted, as the heat contained in these slags is released into the environment during the tapping process without effective utilization. On the other hand, a significant amount of energy is required to reheat the cold materials for the manufacturing of glass and mineral-wool, often necessitating the addition of high-silica materials to facilitate the production process. In this study, an innovative approach is presented to harness the heat present in molten metallurgical slags to directly produce new raw materials for glass and mineral-wool manufacturing while simultaneously reducing energy consumption and mitigating CO_2 emissions. The theoretical amount of waste glass that can be effectively added in the process has been estimated via thermodynamic calculation. Experimental assessments were conducted to examine the impact of high-silica waste materials on the dissolution, melting, and fluidity. Potential recipes for glass and mineral-wool manufacturing were suggested and an assessment of the energy savings, and reduction in CO_2 emissions with manufacturing of glass and mineral-wool using this innovative approach were included. It was determined that the processes could lead to a reduction of over 26kg of CO_2 emissions per 100 kg of new material used, primarily due to the utilization of heat from slag and the accelerated smelting process.

INTRODUCTION

The glass and mineral-wool industries are energy intensive foundation sectors, facing significant challenges in reducing energy consumption and CO_2 emissions (Dewick and Miozzo, 2002; Kiss et al., 2013; Springer and Hasanbeigi, 2017). A significant amount of energy is consumed in raw material preparation and melting, and a great portion of CO_2 emissions is released from carbonate decomposition of raw materials (Deng et al., 2023; Schmitz et al., 2011). The glass and mineral-wool traditionally produced from cold raw materials, incurs high energy consumption, with heating accounting for over 70% of the total energy use. Now low cost, environmentally friendly, energy-saving and increased-recycling are major driving forces for new raw materials. The glass industry endeavours to maximise the recycled glass content, but a significant amount (100 kTs per year in the UK) of poor-quality recycled glass is unsuitable to be used in glass furnace due to issues such as contamination and particle size being too fine, in addition to large number of other wastes such as glass fibre (Iacovidou et al., 2018). Metallurgical slags such as blast furnace (BF) slag and basic oxygen furnace (BOF) slag offer significant environmental benefits over mined raw materials, such as a higher melting rate and provide a carbonate-free source of calcium (Pan et al., 2016).

Molten slag, a high-temperature by-product of the metallurgical industry, holds substantial potential as a valuable resource for the glass and mineral-wool industries. The current focus is largely on investigating BF slag (Oge et al., 2019; Piatak, 2018; Piatak et al., 2015). However, the widespread use of BF slag as a cement raw material has a high utilization rate. Meanwhile, the considerable volume of steelmaking slag generated by the basic oxygen furnace process (BOF) and electric arc furnace (EAF) process remains underutilized. This is attributed, in part, to its low acidity (SiO_2/CaO), leading to high melting temperature and strong crystallization performance that are unfavourable for glass and mineral-wool production. Additionally, the presence of FeO (~25 wt.%) further hampers product quality. The acidity of metallurgical slag is usually lower than 1.0, while the acidity of mineral-wool is required to be more than 1.2, and even higher for glass (Zhao et al., 2019). Then high-silica raw materials are needed to adjust the composition of the metallurgical slags, and high-silica wastes, such as the waste glass become a potentially available resource.

This research aims to use the waste energy during molten slag tapping to convert various hard-to -use wastes into high value, new raw materials for glass or mineral-wool manufacturing. In this work, the energy utilization, and the effective use of waste glass in new materials generation were calculated. Thermodynamic calculations and experiments were carried out to assess the impact of waste glass on molten mixture properties. Modified mixtures were suggested to meet glass and mineral-wool manufacturing requirements. The study also evaluated energy savings and CO_2 emission reductions achieved through this innovative approach.

EXPERIMENTAL

Materials preparation

The BF slag, BOF slag, and low-quality waste glass were sourced from industrial partners. The mineral phases of BOF slag and amorphous phase of BF slag and waste glass were confirmed by X-ray diffraction (XRD) as showed in **Figure 1**. The chemical composition of slags and glass were analysed by X-ray fluorescence (XRF) as presented in **Table 1**. The BOF slag exhibits a high content of FeO and a low acidity, contributing to the facile crystallization of steel slag. The XRD pattern of the BOF slag appears intricate, revealing numerous overlapping peaks indicative of the diverse minerals present in the sample. Iron oxides are prevalent, and beyond the presence of FeO, the XRD pattern suggests the likely existence of brownmillerite ($Ca_2(FeAl)_2O_5$), and magnesioferrite ($MgFe_2O_4$).

TABLE 1 – Composition of BOF slag and waste glass, wt.%

wt.%	CaO	SiO_2	MgO	Al_2O_3	SO_3	P_2O_5	K_2O	TiO_2	MnO	FeO	Na_2O	Acidity
BF slag	41.10	37.01	7.85	11.24	0.91	/	0.58	0.64	0.37	0.35	/	0.90
BOF slag	40.78	16.28	5.57	4.66	0.32	1.68	/	0.64	2.07	27.55	/	0.40
Waste glass	11.25	72.00	1.15	1.50	0.25	/	0.50	/	/	0.05	13.30	6.40

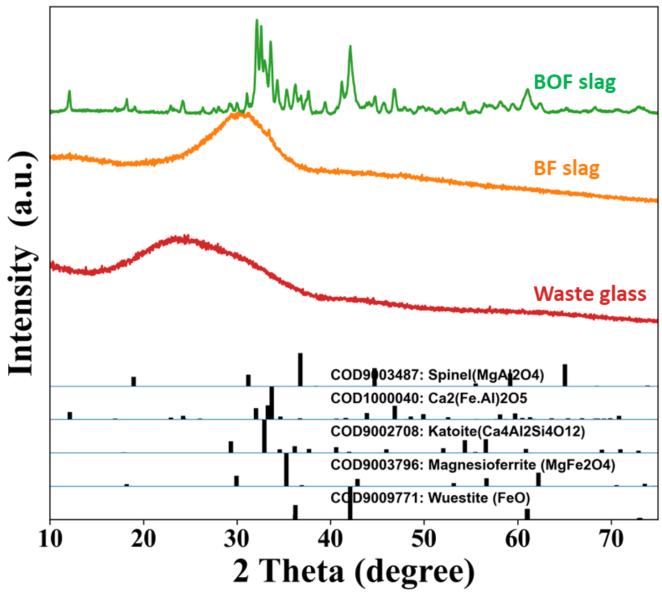

FIG 1 – XRD results of raw materials.

Thermodynamic calculation

The thermodynamic calculation was conducted by FactSage 8.2 using FactPS and FToxid databases (https://www.factsage.com/, 2022). This includes determining the effect of waste glass addition on the temperature and composition of mixture, the equilibrium phases during slag cooling, the melting behaviour and viscosity of raw materials. When calculating the theoretical addition of waste glass into molten slag during tapping, a key consideration is maintaining the slag-glass mixture in a liquid state. The calculations assumed a BF slag tapping temperature of 1480°C and BOF slag tapping temperature of 1550°C. The oxygen potential keeps at 0.21. Adiabatic calculations were also employed to get the temperature after mixing. The viscosity module was used to calculate the viscosity of the slags and molten mixture at liquid stage, with the aim of evaluating the effect of adding waste glass on the fluidity of the mixture.

Waste glass dissolution

The experiments of dissolving waste glass into BF slag were carried out in a high-temperature laser scanning confocal microscope at 1400°C and 1450°C in Argon, the schematic diagram for device and the temperature program are shown in **Figure 2**. The dissolution times were 0s, 10s, 30s, and 60s. The heating and cooling rates were set as quickly as possible at 200°C /min and -300°C /min, respectively. About 100mg of slag was first melted and then cooled down. A glass particle around 5mg was placed on the top of the slag and reheated to the target temperature for a certain time. The cooled samples were analysed using a scanning electron microscope (SEM, Zeiss, Sigma) equipped with energy dispersive X-ray spectroscopy (Oxford, Ultim Extreme).

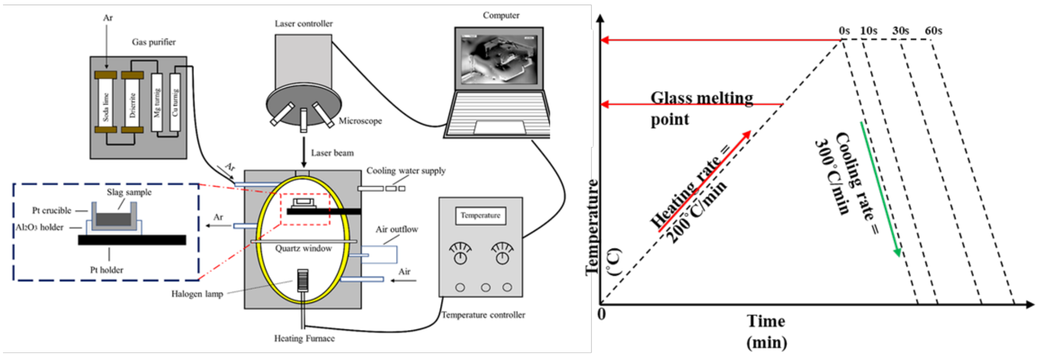

FIG 2 – Waste glass dissolution in BF slag via high-temperature laser scanning confocal microscope.

Melting temperature measurement

Based on the composition range of mineral-wool in the literature, a potential recipe was proposed. A high-temperature vacuum furnace was utilized to determine the melting temperature (T_h) using the Leitz microscope test method. Further information on the equipment and test procedure is available in a previously published paper (He et al., 2023). A cylindrical sample (3 mm in diameter and 3 mm in height) was prepared. During this heating phase, the shape of a compressed cylinder of slag powder was continuously monitored every second with a high-resolution optical camera when the chamber temperature over 1100°C. The heating rate was maintained at 6 °C/min. The melting temperature is defined as the point at which the height of sample is reduced to half of its original size. Additionally, the fluidity temperature (T_f) is determined as the temperature at which the height of sample is reduced to one-quarter of its initial height.

Mineral-wool preparation

Utilizing the proposed recipe, mineral-wool was manufactured through a centrifugal process. The details of the centrifugal apparatus were described in previous paper (He et al., 2023; He et al., 2020). For the process, a crucible containing 200 g of mixture was heated to approximately 1550°C in the induction furnace and maintained at this temperature for about 30 minutes. It was then brought up to a predetermined speed via the rotational drive system. The molten slag was gradually poured from the crucible onto the rotating cup, where it was transformed into wool fibres by centrifugal force. The mineral-wool obtained was observed using a digital optical microscope (OM, Keyence VHX7000).

RESULTS

Thermodynamic calculation

The basic melting characteristics and fluidity of the raw materials can be obtained via thermodynamic calculation, as shown in **Figure 3**. The liquidus temperature of BF slag is approximately 1375°C. After cooling, the predominant phase formed is Mellite. When the temperature drops to 25°C below the liquidus, the solid phase content exceeds 50%. This indicates a significant increase in the solid fraction at relatively small deviations from the liquidus temperature, reflecting the sensitivity of BF slag composition to temperature changes. The melting point of BOF slag is over 1600°C. Owing to its high basicity and FeO content, the spinel phase is precipitate first. Then there is an extensive precipitation of Dicalcium Silicate (C_2S) due to the high basicity. As the temperature falls, there is a rapid reduction in the quantity of the liquid phase, highlighting the sensitivity of BOF slag to temperature changes as well. The glass starts to soften and transition into a semi-molten state at temperatures above 500°C, with its liquidus temperature being approximately 1150°C. At this stage, the wollastonite phase is the first to precipitate. The viscosity of BF slag in its liquid phase is less than 0.6 Pas, demonstrating relatively low resistance to flow. In contrast, BOF slag exhibits a lowe viscosity even solid phases presents, below 2.0 Pas using the Enstein equation (Yue et al., 2018), indicating excellent fluidity. This fluidity is directly influenced by the slag basicity: higher basicity enhances fluidity. This is due to alkaline oxides like CaO, MgO, and FeO disrupting the [SiO_4] network structure in molten slag, which consequently reduces the degree of polymerization of slag (Yan et al., 2019; Yan et al., 2016). As a result, slag with higher basicity presents lower viscosity in its liquid phase. Conversely, glass, with its significantly high SiO_2 content, exhibits a much higher viscosity.

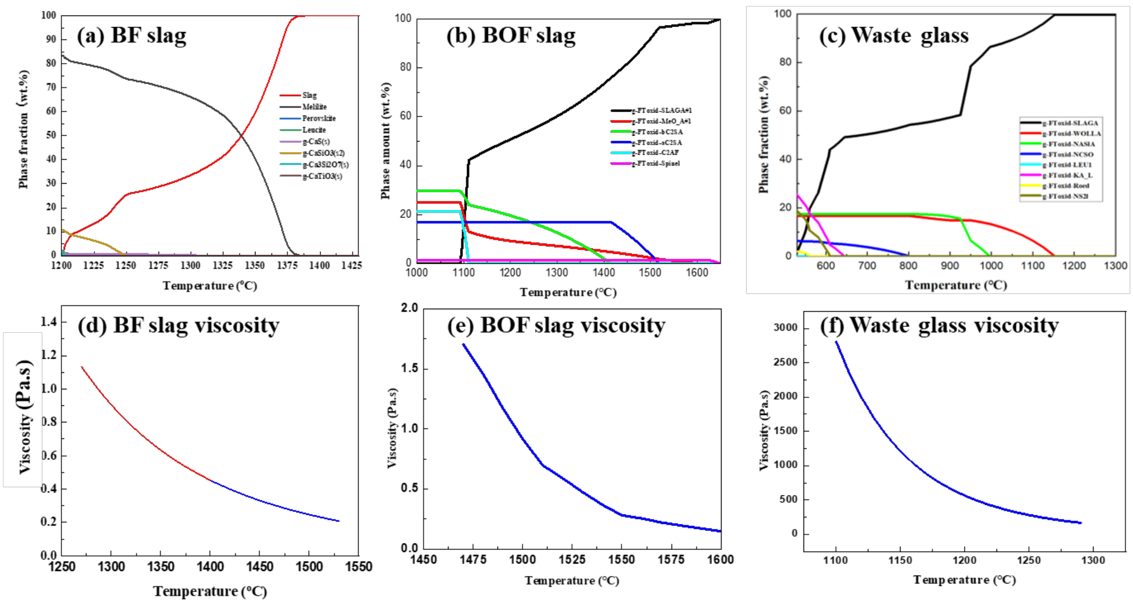

FIG 3 – The equilibrium phases during cooling of raw materials: (a) BF slag, (b) BOF slag, (c)Waste glass, and the viscosity of raw materials obtained via FactSage: (e) BF slag, (f) BOF slag, (g)Waste glass.

Through adiabatic calculations, the impact of waste glass addition on the temperature of BF slag during tapping was assessed, and the melting temperature of these mixtures was also determined. The findings are presented in **Figure 4**. It should be noted that waste glass can introduce a certain amount of carbon, as it may contain organic oil or plastic. The oxidation of this carbon contributes additional heat. The addition of waste glass results in a gradual decrease in the temperature when waste glass containing 0% carbon and more than 9.4 kg of waste glass led to a temperature less than the liquids (**Figure 4a**). Conversely, adding more than 50kg of waste glass with 5% carbon into slag content keeps the mixture in a liquid state (**Figure 4b**). As shown in **Figure 4c**, when the temperature is reduced to 1350°C, the viscosity of the mixture with 30% added glass remains below 2 Pa·s, maintaining a good fluidity. For BOF slag, due to the oxidation of FeO and 0% carbon, the slag temperature under adiabatic conditions will exceed 2000°C, as shown in **Figure 5.** The temperature of the mixture is still higher than the melting temperature when the amount of waste glass added exceeds 50kg. It is worth noting that heat loss is not considered here. Since the BOF slag possesses a higher basicity compared to the BF slag, its viscosity is more favourable when the same quantity of waste glass is added. Consequently, maintaining fluidity is less challenging with BOF slag.

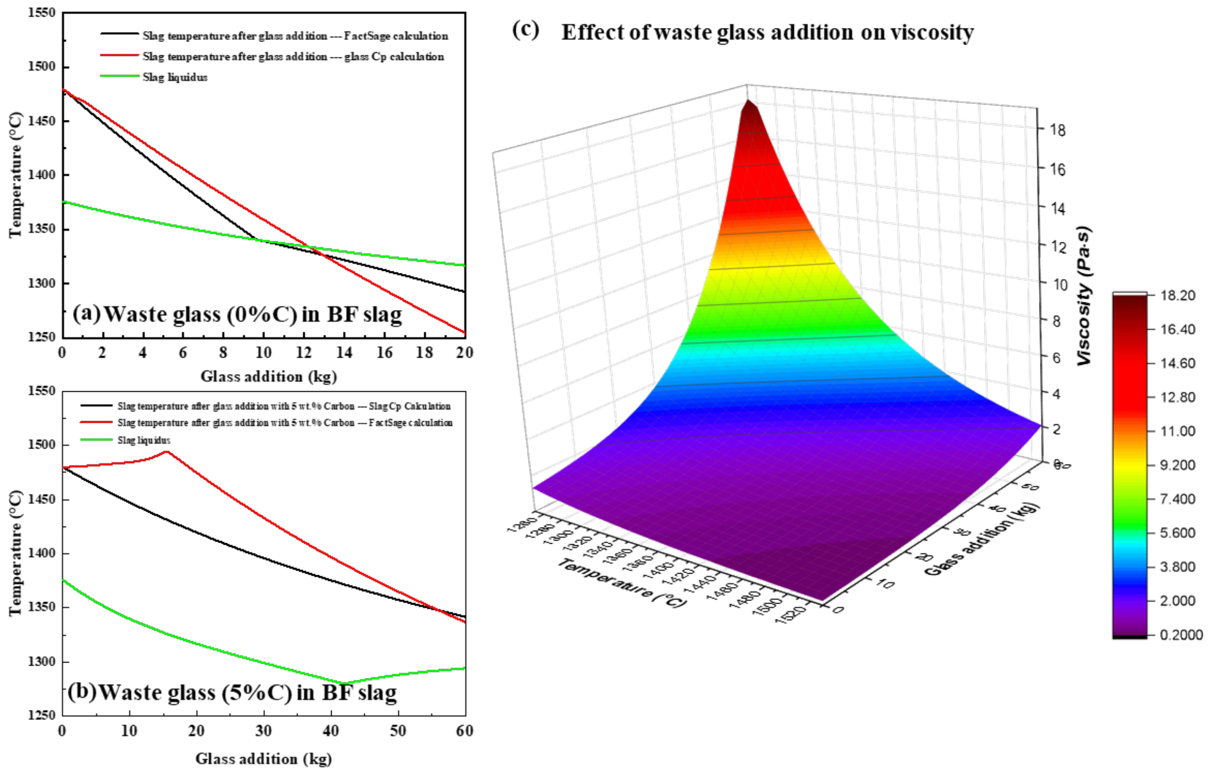

FIG 4 – The theoretical addition of waste glass with different carbon content into BF slag during tapping: (a) 0% Carbon, (b) 5% carbon, and the effect of temperature and waste glass addition on the viscosity.

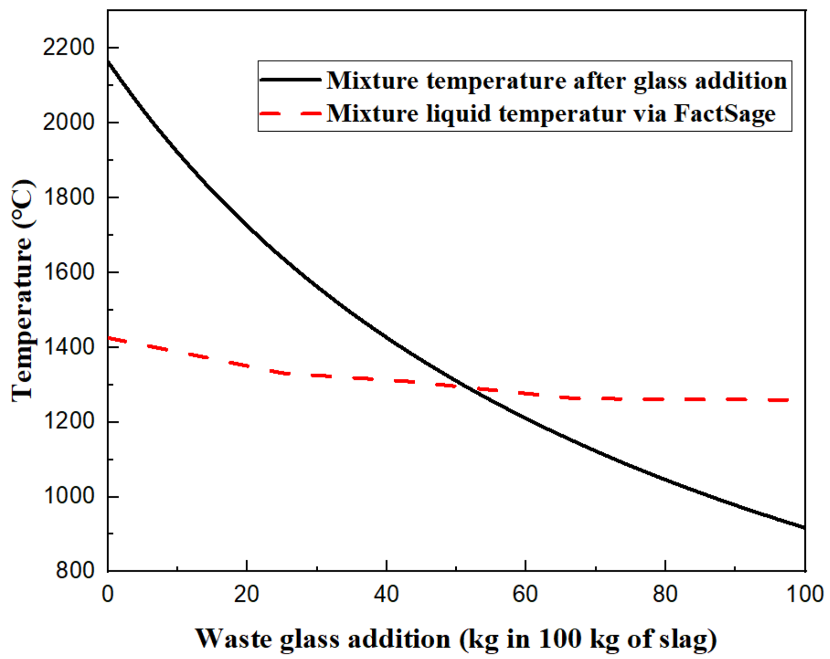

FIG 5 – The theoretical addition of waste glass with 0% carbon into BOF slag during tapping.

Dissolution of waste glass in molten slag

Figure 6 displays the SEM-EDS results illustrating the cross-section microstructure of waste glass dissolution in BF slag at 1400°C and 1450°C for 60 seconds. The top surface layer of the slag is identified as glass, characterized by a high concentration of sodium and silicon, as revealed by EDS analysis in **Figure 6a**. At a temperature of 1400°C, the glass has completely melted, and the liquid glass layer begins to dissolve, but the dissolution is not complete. Remarkably, when the temperature is increased to 1450°C, the glass layer in the BF slag can be completely dissolved in less than 60 second. Since no apparent stratification is evident and the distribution of sodium is uniform throughout, it indicates a homogenous composition in the slag layer. Due to the high SiO_2 content in waste glass, a lower SiO_2 concentration in the slag leads to a stronger driving force for dissolution, accelerating the dissolution rate. Additionally, SiO_2 dissolution is faster at higher temperatures and lower viscosities. Consequently, at same temperature, waste glass dissolves faster in BOF slag compared to BF slag. The melting and dissolution processes are further enhanced by stirring during the addition of waste glass.

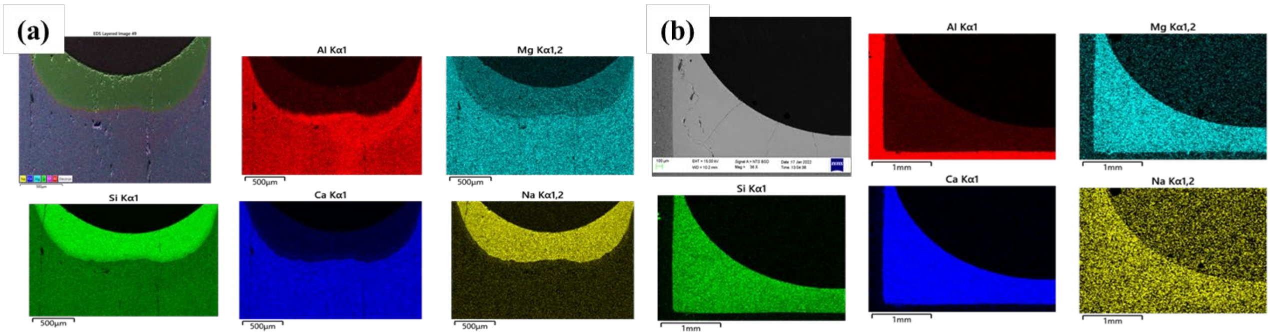

FIG 6 – Dissolution of waste glass in BF slag at different temperature for 60s: (a) 1400°C, and (b) 1450°C.

DISCUSSION

Integrating hard-to-use waste glass into the metallurgical slag during the tapping process promises to achieve the multiple purposes of waste utilization, energy saving and waste heat recovery, as schematically illustrated in **Figure 7**. This process utilizes the sensible heat of metallurgical slag and the heat of reaction from the oxidation of iron and organics, mixing hard-to-use high-silica waste with molten slag to form new raw materials suitable for glass manufacturing or mineral-wool production. The application of the new material after mixing is determined based on the compositional requirements of glass and mineral-wool. According to thermodynamic calculations of melting performance, fluidity, and solubility experiment results, for BF slag, if there is 5% carbon in the waste glass, the addition of waste glass is about 30% which still meets the requirements for slag transportation and tapping. For BOF slag, the addition of waste glass can exceed 50% due to the high tapping temperature, high basicity and high FeO content. Depending on the composition of the new material and the final composition of the expected products, glass or mineral wool is prepared by adding other raw materials. Generally speaking, the new material obtained by BF is suitable for glass manufacturing, while the new material obtained by BOF is suitable for mineral wool. Two common approaches to utilizing these materials will be explored below.

Based on a typical container glass composition (Shelby, 2020), a potential formula has been proposed as shown in **Table 2**. This recipe allows for the use of only 20% new raw materials in the manufacturing of container glass. As a result, about 30% of waste glass and 15% BF slag are used in this glass production. By incorporating this new material, there is a notable reduction in lime consumption, which consequently leads to a decrease in CO_2 emissions. The new raw material contains sulfur which is a powerful refining agent and benefits glass manufacturing (Hujova and Vernerova, 2017). Additionally, the new material serves as a replacement for nepheline syenite and feldspar, which are the primary sources of alumina in traditional glassmaking. This substitution not only utilizes waste materials but also maintains the necessary chemical composition for high-quality glass production.

FIG 7- Conversion of wastes to new raw materials for glass and mineral-wool manufacturing

TABLE 2 – A potential recipe for soda-lime-silica (container glass) using new material, wt.%

	Ratio	CaO	SiO_2	MgO	Al_2O_3	SO_3	K_2O	TiO_2	MnO	Fe_2O_3	Na_2O
New material 1 50kg waste glass in 100kg BF slag	20	32.12	47.58	5.84	8.31	0.71	0.56	0.45	0.26	0.28	4.00
SiO_2 sand	46	0.28	98.2	0.03	0.28	0.07	0.01	0	0	0	0.03
Soda ash	16	0	0	0	0	0	0	0	0	0	58.49
Waste glass (good quality)	23	11.25	72	1.15	1.5	0.25	0.5	0	0	0.05	13.3
Lime	2	92.01	1.51	0	0.93	0	0	0	0	0.11	0.00
Proposed glass composition		10.67	70.78	1.30	1.96	0.22	0.22	0.08	0.05	0.06	14.04
Typical container glass composition		10-12	70-73	0.5-2	0-2	0-0.5	0-1	0-0.1	/	0-1	12-15

TABLE 3– A potential recipe for mineral-wool using new material, wt.%

	Ratio	CaO	SiO_2	MgO	Al_2O_3	K_2O	TiO_2	MnO	Fe_2O_3	Na_2O
New material 2 50kg waste glass in 100kg BOF slag	60	25.8	45.45	3.2	3.48	0.25	0.33	1.06	14.57	4.66
SiO_2 sand	15	0.28	98.2	0.03	0.28	0.01	0	0	0	0.03
Bauxite	15	0	18.68	0	56.29	0	2.33	0	1.29	0
Dolime	10	54.75	3.55	40.1	0.33	0.08	0.05	0.03	0.38	0.02
Proposed material composition		21.00	45.16	5.93	10.61	0.16	0.55	0.64	8.97	2.80
Mineral-wool composition from literature		10-25	41-53	6-16	6-14	0-2	0-3.5	0-1.1	0-13	0-6

Referring to the composition ranges of mineral-wool documented in the literature (Yliniemi et al., 2021), a potential recipe was proposed, as shown in **Table 3**. This formulation permits the incorporation of approximately 60% new raw materials. Consequently, in producing this specific type of mineral-wool, around 20% of the raw materials are derived from waste glass and another 40% from BOF slag. In the laboratory experiments, analytically pure reagents (SiO_2, Al_2O_3, and MgO) were used for replacing silica sand, bauxite, and dolime. These were mixed with BOF slag and waste glass to create a mixture with a similar composition to the recipe. Then this mixture was used for both the measurement of the melting temperature and the preparation of mineral-wool, and the results are presented in **Figure 8**. The Lab-made mixture shows a melting temperature at approximately 1160°C, and its fluidity temperature is around 1225°C. The diameter of fibres is a crucial determinant of the properties of mineral-wool products, affecting characteristics such as chemical stability and sound absorption (He et al., 2023; Liu et al., 2013). The mineral-wool produced has fibres measuring approximately 10-20 um in diameter, which are over twice as thick as those found in commercial rock wool products. A high-speed, four-roller centrifuge will be utilized to produce wool fibres and will be investigated in future work.

When the raw materials are used directly in their molten state for production, it can significantly enhance the melting rates, reduce the batch-free times, and decrease the bubble content in the final product. In this case, the heat from slag could be recovered directly. By directly using the sensible heat and exothermic reactions of slag, there is a potential energy saving of 0.16 GJ per 100 kg of new material with 1350°C used. This translates to a reduction in standard coal consumption by 7 kg and cuts carbon dioxide emissions by 26 kg. When considering the energy used for melting natural raw materials, the reduction in CO_2 emissions is further amplified by replacing natural raw material with these new raw materials. This approach not only contributes to lowering greenhouse gas emissions but also promotes the recycling of waste glass, aligning with sustainable waste management practices and environmental conservation efforts.

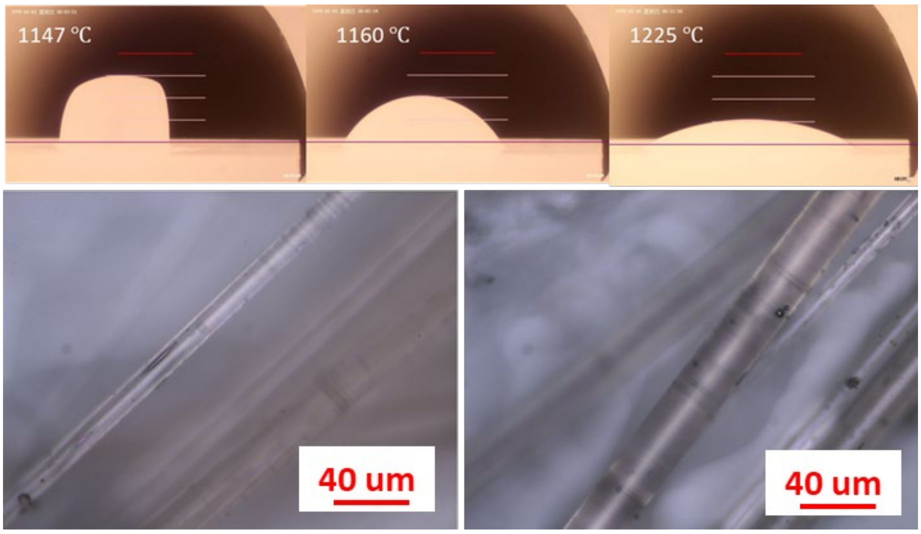

FIG 8- Height changes of Lab sample (a), and the morphology of obtained mineral-wool (b).

CONCLUSIONS

1. This study introduces an innovative approach for recovering the heat from high-temperature ironmaking and steelmaking slags, which are typically discarded and release heat into the environment during the tapping process. This heat is instead utilized to recycle hard-to-use waste glass in producing raw materials directly for glass and mineral-wool manufacturing.
2. Thermodynamic assessments indicate that for BF slag with 5% carbon in waste glass, up to 30% waste glass can be added without affecting transport and tapping requirements. For BOF slag, over 30% waste glass even with 0% carbon content addition is feasible owing to its higher tapping temperature, basicity, and FeO content.

3. New recipes for container glass and mineral-wool have been suggested, based on the typical products composition ranges. Utilizing the sensible heat and reaction heat of slag could reduce coal consumption by 7 kg and CO_2 emissions by 26 kg for every 100 kg of the new material.

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