CERAMIC FRIT MELTING BY ELECTROMAGNETIC INDUCTION

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ABSTRACT

The ceramic tile manufacturing industry faces the challenge of decarbonising its production processes to reduce CO_2 emissions, chiefly from the heat treatment stages. Within the ceramic process, one of the most energy-intensive stages is the melting stage to obtain the ceramic frits used in the preparation of glazes, which takes place in continuous melting furnaces at temperatures of around 1500ºC. The frit precursor material in these furnaces is a homogeneous mixture of different raw materials in powder form that is fed continuously into the melting chamber. As the material melts, it is evacuated from the chamber by a heated chute that pours the melt into a refrigerated water bath where it is quenched, thus producing the glassy material that constitutes the frit. The heat for these furnaces comes mainly from natural gas combustion, using air or oxygen as the combustion agent.

Electrification is a promising alternative in the decarbonisation of this process and can be implemented in different ways, such as heating by the Joule effect using electrical resistors, generating heat within the material to be melted from the effect of a microwave field, using electrodes immersed in the melt, or heating by electromagnetic induction. At present, there is no specific electrical equipment on the market for melting ceramic frits using any of these technologies.

Electromagnetic induction is widely used in the melting of metals, but it is complex to apply in the manufacture of vitreous materials such as frits, due to the high electrical resistivity of the raw materials used to produce frits, which impedes the induction of electrical currents in the material that would allow its heating. This research presents a trial of a melting concept for obtaining ceramic frits by electromagnetic induction, with a view to electrifying the manufacturing process at pilot scale using clean energy, low emissions and strict process control.

1. INTRODUCTION

The European Green Deal aims to achieve greenhouse gas emission neutrality by 2050. The decarbonisation of energy-intensive industries is essential to achieving that goal [1].

The roadmap towards successfully attaining that objective includes measures such as:

- Reduction of CO_2 emissions, by selecting raw materials and/or increasing energy efficiency.
- Switching to renewable energy sources (green hydrogen, biofuels, green electricity).
- CO_2 capture.
- Other carbon removal technologies.

In the glassmaking process, whether the glass be flat, hollow, domestic, filament, mineral wool, etc., the most common and conventional way to provide heat to melt the glass is by burning fossil fuels. Sometimes, this primary energy input from combustion is backed up by electrical energy through electrodes immersed in the melt. This technique improves the energy transfer in the melt and is commonly known as "boosting".

According to the literature [1-2], electric furnaces may be used in specific applications, such as the production of high-temperature insulating glass wool or in small-volume furnaces for specialty glass. In small-sized furnaces, the efficiency of burning fossil fuels decreases because the thermal losses per ton of molten material are significantly higher. Thanks to electricity's more efficient heating and higher melting capability per unit area, the difference in cost of electric power compared to that of fossil fuels is offset in small furnaces. However, in order for the electric current to flow through the glass and heat it, the glass must first be melted, which implies using fossil fuels at the start of the melting process. In addition, these furnaces have a relatively short life, ranging from 2 to 7 years depending on the type of glass produced and the operating conditions. In any case, examples of electrical heating for glass manufacturing are not widespread [2-3].

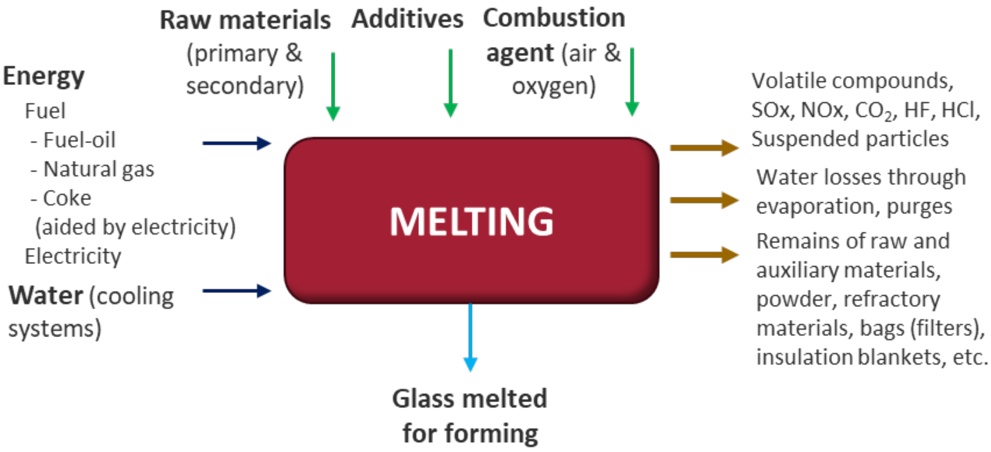

Figure 1. *Environmental considerations in the melting stage of glassmaking [3].*

The furnaces used to manufacture ceramic frits are significantly smaller than those used to produce glass. Currently, the production capacity of most furnaces does not exceed 20 tons per day. In these furnaces, the heat source is generated by burning natural gas using air or oxygen as the combustion agent. To date, there are no known experiments on an industrial scale of using electric melting furnaces - their use is limited to laboratory furnaces.

Electrification is emerging as a promising alternative for the decarbonisation of the melting process, and may involve different heating techniques, such as heating with electrical resistors, applying microwaves, immersed electrodes, electromagnetic induction heating, or a combination of those techniques.

Although electromagnetic induction is widely used in metal melting, its application in the manufacture of glassy materials such as frits is more complex, due to the high electrical resistivity of the raw materials in solid state, which prevents the induction of electrical heating currents within the material. Nevertheless, the advantages this type of heating offers compared to other methods makes it worthy of further exploration. Such advantages include high productivity due to direct heating of the material, high energy efficiency, ease with which temperature control can be automated through a closed loop, preservation of product quality by avoiding contact with flames or foreign bodies, and the possibility of using sustainable energy sources.

Induction heating involves the generation of strong magnetic fields that are used to induce currents in a piece of conductive material. The coil that generates these magnetic fields is called an inductor. In electromagnetic induction heating, mains AC is converted into direct current by a current inverter before being transformed by an oscillator into alternating current at the desired frequency and applied to the inductor. The magnetic field generated in the inductor induces current in the workpiece and thus heats it, as long as the material has low resistivity, i.e., is electrically conductive [4].

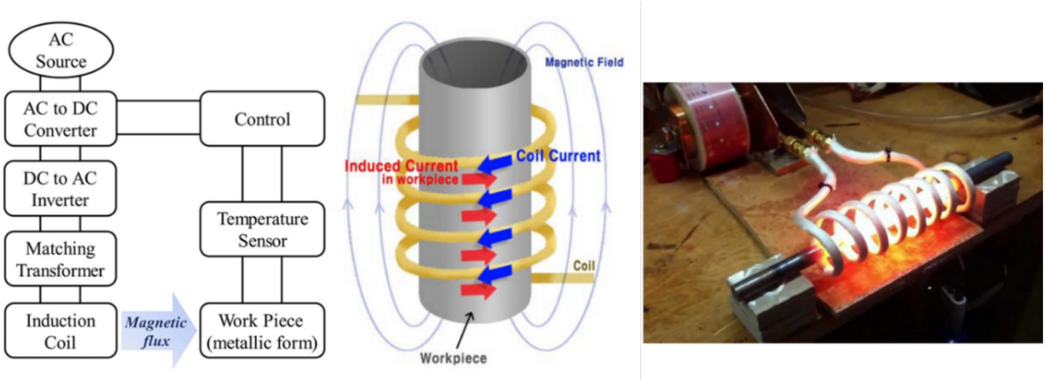

Figure 2. a) *Principle of induction heating. b) Induction heating of conductive material.*

Material melting techniques based on the application of electromagnetic induction are divided into two main categories: indirect heating and direct heating. In the former, the material is melted by the heat generated by auxiliary elements that reach high temperatures thanks to electromagnetic induction. On the other hand, in the second group, the actual material to be processed (charge) heats up because of its susceptibility to the electromagnetic field generated.

Within direct melting techniques, a distinction can be drawn between the hot crucible method, where melting occurs in a refractory crucible heated by the molten material, and the cold crucible method, in which melting takes place in a metal crucible, usually copper, which is continuously cooled. This creates a thin layer of solidified glass between the crucible and the molten charge, which acts as a thermal insulator, preventing the crucible from melting. The latter method is more expensive than the former and is used only in situations where it is crucial to avoid contamination of the molten material, such as in titanium melting processes [5].

In order to achieve a continuous melting process to manufacture frits by induction heating, it is essential to overcome a crucial initial hurdle: the electrically insulating nature of the ceramic powder material at room temperature. That means that electromagnetic induction alone is not capable of directly heating the frit precursor powder material. However, as reported in the literature, certain glass-forming compositions can become sufficiently conductive once melted, allowing them to be heated using the technique at certain working frequencies in the inductor [6]. The implication is that the initial heating of the powder needs to be carried out indirectly, using electrically conductive susceptors that are heated by induction and then transfer the heat to the powder, causing it to melt.

2. OBJECTIVE

This study focuses on assessing the technical feasibility of melting ceramic frits by direct heating using electromagnetic induction. The ultimate goal is to electrify the manufacturing process on a pilot scale, with clean energy, minimal emissions, and a high standard of process control.

3. MATERIALS AND EQUIPMENT

3.1. COMPOSITIONS STUDIED

Table 1 shows the chemical composition of the main oxides in three commercial frits selected for the study. The choice was made on the basis of significant differences in their fusibility and alkali and alkaline-earth element content. The melting temperature of the three compositions studied was measured by hot stage microscopy, and revealed a melting temperature of 1060ºC for composition A, 1165ºC for composition B, and 1385ºC for composition C.

Oxide	A (%)	B (%)	C (%)
SiO_2	51-55	57-61	62-66
Al_2O_3	11-15	4-8	7-11
B_2O_3	5-7	2-4	8-10
R_2O^*	19	6	8
RO^{**}	3	17	6
ZrO_2	6	0	0
ZnO	0	10	4

* Na_2O and K_2O. ** CaO, MgO and BaO.

Table 1. *Chemical composition in oxides of the studied frits (% by weight).*

3.2. SYSTEM FOR MEASURING THE ELECTRICAL PROPERTIES OF MELTS

The ability of a molten material to be heated by electromagnetic induction is closely related to its electrical resistivity [6]. To measure this electrical parameter, a low-precision measurement method based on the conduction of an electric current between two electrodes immersed in the melt has been used to determine the material's electrical impedance [7] [8] [9]. To convert the electrical impedance measurement into electrical resistivity, the measuring cell must first be calibrated. This calibration process was carried out using potassium chloride solutions of analytical quality as standards and known conductivities [10].

Figure 3. *Detail of the crucible and measuring electrodes used to characterise the electrical properties of the melts.*

3.3. ELECTROMAGNETIC INDUCTION FURNACE

For this research into frit melting processes using electromagnetic induction, a specific prototype was developed and built to assess melting with this technique (Figure 4).

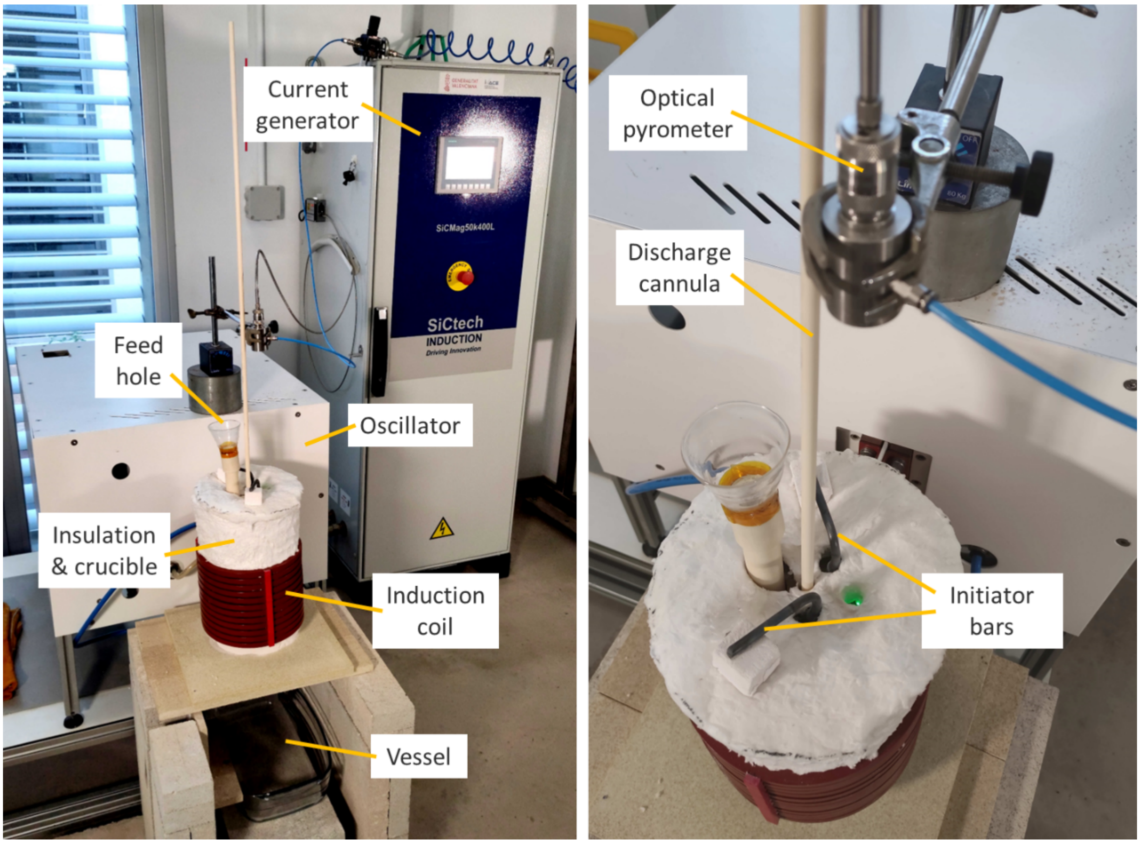

Figure 4. *Overview and detail of the prototype developed to carry out research on melting ceramic frits by electromagnetic induction.*

The main control panel contains part of the electrical installation, the PLC controlling the equipment, and the current generator. The current generated is transformed in the oscillator and applied to the inductor coil generating the magnetic field. To maintain the required temperature, the whole system is water-cooled through a closed loop system.

An alumina crucible is placed inside the inductor coil together with a high temperature fibre insulator. The experimental set-up is completed with a refractory cover that houses the melting-initiator bars, an opening through which the powder to be melted is fed, an optical pyrometer to measure the temperature of the material, and a cannula that acts as a discharge valve to control the pouring of the melted material into a water bath through a hole drilled in the bottom of the crucible.

4. RESULTS

4.1. MEASUREMENT OF MELT ELECTRICAL PROPERTIES

To determine the electrical resistivity of the melts, a small alumina crucible is filled with the frit precursor powder sample to be analysed. The crucible is then placed inside a high-temperature furnace with the electrodes positioned close to the surface of the sample although not physically touching it. Once the desired temperature is reached, the electrodes are slowly lowered until an impedance reading is taken. At that point, the electrodes are inserted further into the melt and the insertion distance is taken as a reference in the successive tests carried out with the various samples.

With the electrodes correctly positioned, a sweep of impedance measurements is carried out at different frequencies and melt temperatures. Figure 5 illustrates the electrical impedance measurements made for sample B at three melt temperatures. It is clearly seen that the electrical impedance measurement is significantly influenced by temperature. As temperature increases, ionic mobility is incremented and electrical impedance diminishes [7].

These electrical impedance measurements were performed for the three frit samples at three different melting temperatures. Figure 5 shows the results of transforming the impedance measurement to electrical resistivity for the three samples. Electrical resistivity is seen to have an Arrhenius-type relationship with temperature while, additionally, the significant disparity in resistivity values depending on the sample analysed is to be highlighted.

Sample A is the only sample that returns resistivity values below 10 Ω·cm and low sensitivity with temperature. This could be due to its lower melting temperature and its composition rich in alkaline elements, which have a smaller ionic radius and, therefore, higher ionic mobility when subjected to an electric field [9].

In contrast, sample B exhibits the highest electrical resistivity of all samples analysed and high temperature-dependence. This sample has a higher melting point and is rich in alkaline earth elements and zinc, ions with a larger ionic radius [11].

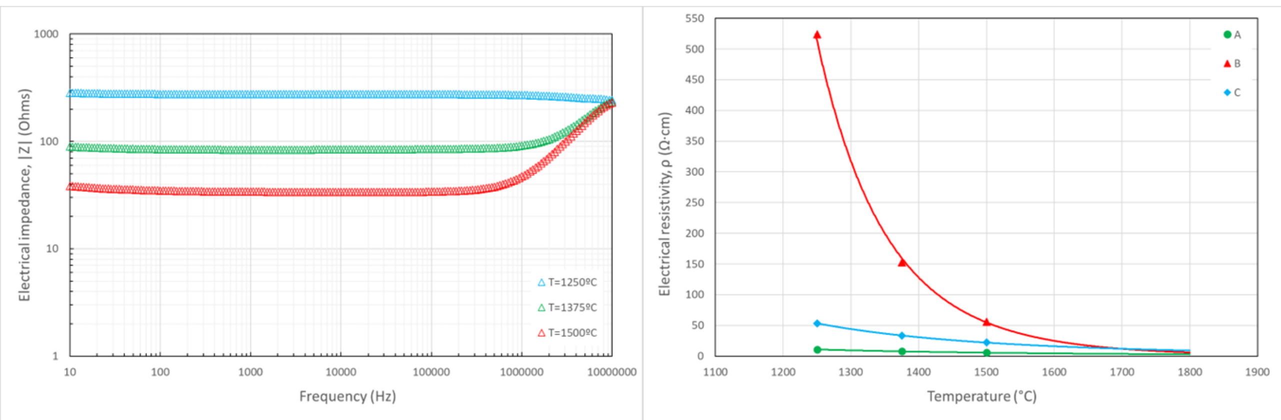

Figure 5. *Electrical impedance versus measurement frequency for sample B at three temperatures (left). Electrical resistivity versus temperature for samples A, B and C (right).*

4.2. ELECTROMAGNETIC INDUCTION MELTING TESTS

4.2.1. STATIC MELTING TESTS

Initially, the tests were focused on verifying whether the ceramic frit melting process could be carried out by electromagnetic induction once the precursor powder had melted and the initiator bars had been removed. Figure 6 shows the evolution of material temperature and current intensity applied during a temperature holding test on frit A. The crucible was charged with 350 grams of material, and power was gradually applied to avoid crucible breakage until 1100°C was reached. At that point, more material was added to the crucible to bring total mass up to 750 grams. The temperature reading of the material was seen to decrease with each addition, due to the fact that part of the added powder was placed under the inspection hole (1). When 1400°C was reached, the initiator bars were removed (2), leading to a drop in temperature, which was offset by increasing the power applied. To confirm definitively that the molten frit was capable of being heated and melted by induction, two further amounts of 100 grams of material were added, and the temperature was seen to recover (3). When 1400°C was reached again, induction was interrupted (4). The crucible was allowed to cool down and, when it reached 1100°C, induction was reactivated (5) before the frit solidified. The temperature of the material was then seen to progressively increase (6). This experiment proved that, once the frit was in a liquid state, the composition could be heated by induction without any need for an external auxiliary element or source.

Figure 7 shows the temperature holding tests for frits B and C performed after the bars were removed. In these tests, a larger initial amount of material was charged and more material (1) was only added once to reach a total of 750g, as in the previous test. Power was applied gradually until the frit reached 1500°C, at which point the bars were removed (2). Despite power being increased when the bars were removed, melt temperature was seen to decrease rapidly. The conclusion is that, with these frit compositions at the present reactive power and working frequency, it is not possible to maintain the melt without the aid of an auxiliary element.

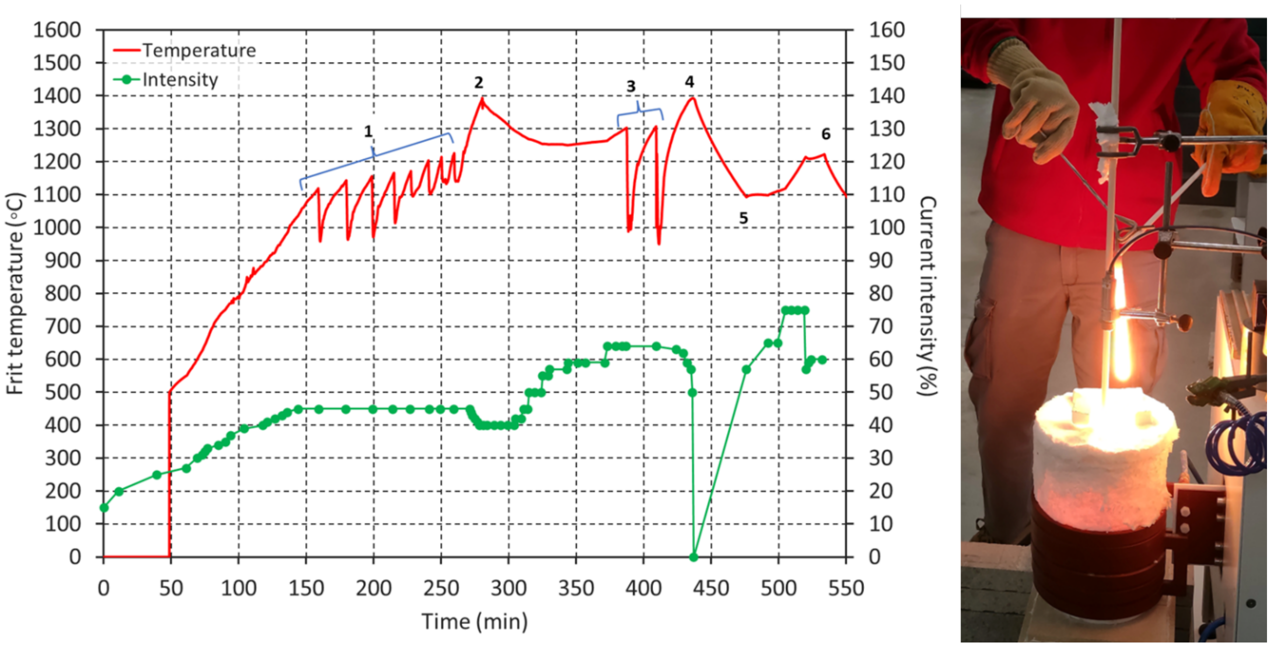

Figure 6. *Evolution over time of the most significant parameters during a frit A melting test (left). Detail of the initiator bars being extracted (right).*

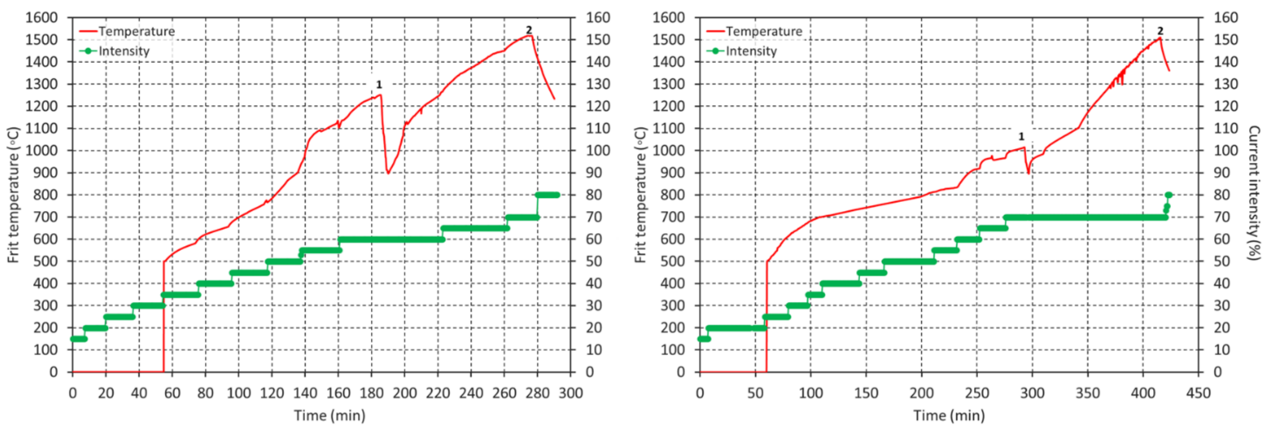

Figure 7. *Evolution over time of the process variables during melting tests of frits B (left) and C (right).*

4.2.2. DYNAMIC MELTING TESTS

In addition to the temperature holding tests, fritting tests were also carried out using the induction technique through the orifice at the base of the crucible. This orifice, plugged by the cannula during heating, allows the frit to flow into the lower vessel filled with water, causing the melt to quench and thus obtaining a vitreous material. Figure 8 shows the heating and fritting test for sample A. The crucible was charged with an initial mass and, when it reached 1000ºC, several additions of material were made to reach a total of 750g (1).

Melt temperature was then increased to over 1300°C, the initiator bars were removed (2), and the power adjusted to maintain a temperature of 1300°C (3). At that point, the cannula plugging the orifice was removed but discharge did not occur due to the melt's high viscosity. Therefore, power was increased to reach a temperature of 1400ºC, at which point the orifice was reopened (4). At that temperature, melt viscosity had decreased sufficiently to obtain a frit, albeit discontinuously, so that the temperature was increased to 1500ºC (5). At that temperature, continuous fritting was achieved until the crucible emptied completely. A ceramic control of the resulting frit was carried out by comparing it with the commercial frit produced from the same composition, and test frit properties and finish were found to be the same.

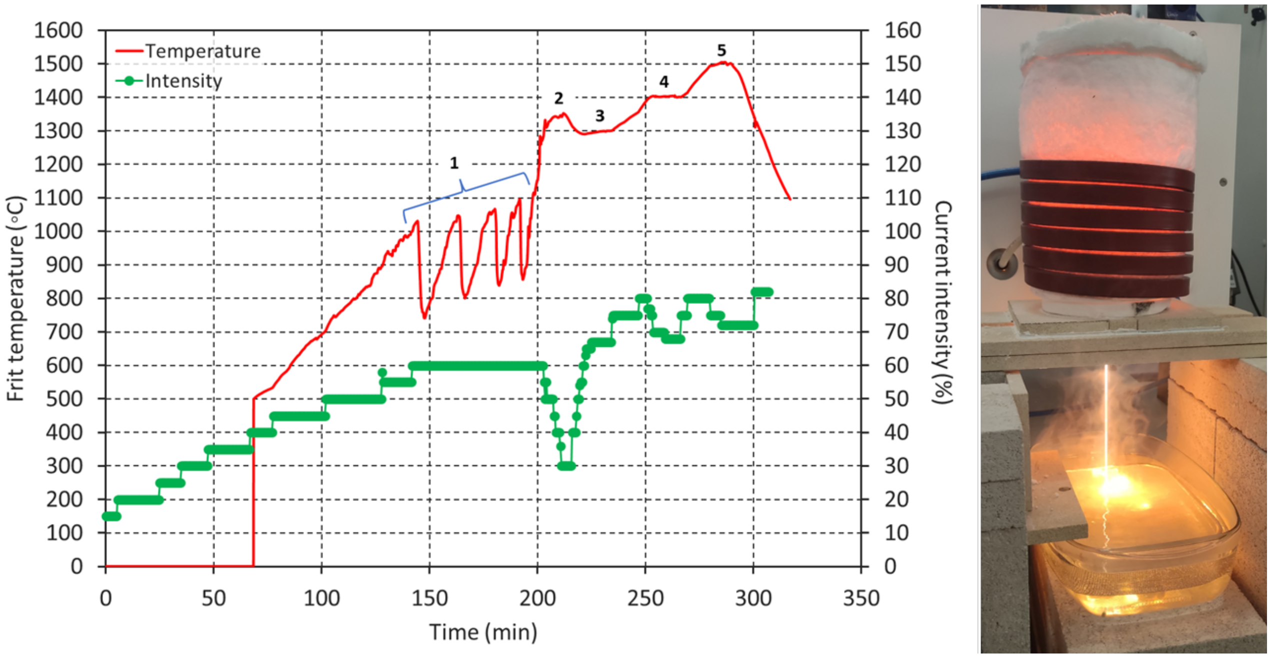

Figure 8. *Continuous melting and fritting test on frit A.*

5. CONCLUSIONS AND FUTURE WORK

The following conclusions can be drawn from this study:

- Decarbonisation of energy-intensive industries is fundamental to achieving the goal of the European Green Deal. The electrification of thermal processes can play a crucial role in achieving that goal.
- Three different frit compositions were successfully melted using the electromagnetic induction technique in combination with melt initiators. However, when the initiators were removed, only one of the three compositions could be kept hot by induction.
- A direct relationship was observed between the electrical resistivity of the sample to be heated and its ability to retain and capture heat on its own once in the liquid state.
- Electrical resistivity is a parameter that varies as a function of temperature and chemical composition of the molten material.

The findings of this study suggest that electromagnetic induction could be considered a promising technique for the direct melting of ceramic frits. However, the preliminary tests indicate that its feasibility depends critically on the chemical composition of each sample, which in turn determines its electrical resistivity at melting temperatures. To determine the limits and real possibilities of this technology, further research will be necessary.

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6. REFERENCES

[1] Draft 1 of the Best Available Techniques (BAT) Reference Document for the Ceramic Manufacturing Industry (CER BREF), European Commission, 2023, JRC134517. https://eippcb.jrc.ec.europa.eu/reference/ceramic-manufacturing-industry

[2] Best Available Techniques (BAT) Reference Document for the Manufacture of Glass (GLS BREF), European Commission, 2013, JRC78091. https://eippcb.jrc.ec.europa.eu/es/reference/manufacture-glass-0

[3] Guía de Mejores Técnicas Disponibles en España del Sector de la Fabricación del Vidrio. Ministerio de Medio Ambiente, 2007, I.S.B.N.: 978-84-8320-389-7.

[4] Richard E. Haimbaugh, Induction Heat Treating 2nd edition, ASM International, Ohio, 2015, ISBN-13: 978-1-62708-089-7.

[5] Saito, K., Okumura, T., & Yamamoto, K. (2015, October). Large Scale cold crucible levitation melting furnace for titanium. In 8th International Conference on Electromagnetic Processing of Materials.

[6] Yu.S. Kuz'minov, E.E. Lomonova and V.V. Osiko, Cambridge International Science Publishing Ltd, Cambridge, 2003, ISBN 978-1-904602-01-9.

[7] Susan L. Schiefelbein, A new technique to measure the electrical properties of molten oxides, Massachusetts Institute of Technology, Massachusetts, 1996.

[8] F. Soleimani et al., Electrical Resistivity Measurement of the Molten Cordierite Glass Using Two wire Method, Advanced Ceramics Progress, ACERP: Vol. 2, No. 1, (Winter 2016) 1-4.

[9] Yusuke Harada et al., Impedance Measurements and Equivalent Circuit Analysis of Binary Alkali Silicate Melts, Department of Materials Science and Engineering, Kyushu University, Japan, 2018, ISIJ International, Vol. 59 (2019), No. 3, pp. 421-426.

[10] G. J. Janz and R. P. T. Tomkins, Conductance Cell Calibrations, 1977 J. Electrochem. Soc. 124 55C.

[11] Yusuke Harada et al., Structural Evaluation of Silicate Melts by Performing Impedance Measurements and Quasichemical Model Calculations, METALLURGICAL AND MATERIALS TRANSACTIONS, 968—VOLUME 52B, APRIL 2021.