STUDY OF MELTING DYNAMICS OF OXIDES IN INDUCTOR CRUCIBLE

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ABSTRACT. Numerical modelling of high frequency inductive heating of oxides with high processing temperatures in inductor-crucible furnace is considered. As particular examples melting of oxides with high melting temperatures and amorphous materials are discussed. Necessary initial heating of the load is performed either by insertion of Mo ring or additional gas burner at the top of the furnace. Either SIMPLER method or direct solution of flow equations are used to account the convection of incompresible melt. Different regimes of voltage, current, and gas burner during a set of stages of induction process are studied. The shrinkage of porous material is included. Basing on the numerical studies, the characteristic power requirements and main heat losses are estimated. Moreover, optimisation of furnace parameters is performed.

INTRODUCTION

High frequency (>200 kHz) skull melting in inductor crucible furnace (ICF) with skull layer as an insulator is well suited for treatment of oxide materials with low heat and electrical conductivities at room temperature and high melting temperature (over 2500 °C). Major heat losses and, consequently, power requirements depend significantly on the properties of this layer [1]. An examples could be oxides like Y_2O_3 stabilised ZrO_2, $ZrSiO_4$, amorphous glasses, etc. The main operational problem is energy transfer to melt at low temperatures, that requires special regime of pre-heating. There are several possibilities of initial heating, e.g.: heating by Mo ring oxidising at high temperatures, heating by gas burner and others. The appropriate control of power is essentially required in all the cases ensuring the heating up to the processing temperatures [1] (for usual oxides above melting temperature, and for amorphous glasses – temperatures with sufficiently low viscosity). When the average load temperature reaches the processing temperature, the melted material in method of skull melting is separated only by thin layer (skull) of the same oxide material with initial porosity.

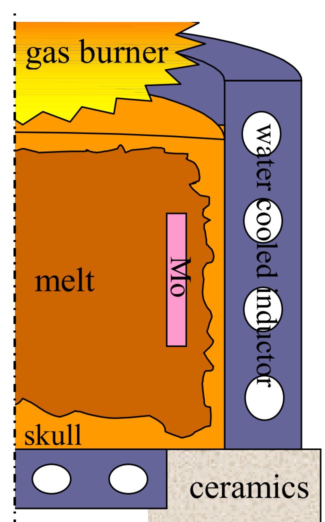

Figure 1. Axial cross-section of furnace for melting of oxides

The characteristic shape of the furnace for melting of zirconium oxide and glass are shown in Figure 1.The massive and water-cooled inductor made either from Cu or Al, which acts

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also as the crucible. The typical values of frequency used for melting of oxides with poor conductivity are several hundreds of kHz. In these simulations the frequency is set to 350 kHz. The cylindrical, metallic ring for initial heating is placed close to the inner surface of inductor in case of zirconium oxide. Numerical modelling of the melting process is built to obtain the most appropriate regime of heating, understanding the process, and optimise the furnace and heating parameters. The system is assumed as axis-symmetrical despite the geometry of the inductor and thermal boundary conditions are not perfectly axis-symmetric because of power supply connection and angularly sectioned cooling system. Axial-symmetric model of calculations has been made where the different possibilities of initial heating are examined. Distributions of electromagnetic, temperature, and velocity fields are closely related and they are calculated simultaneously, because oxide material has exponential dependence of conductivity on temperature and convective heat transfer is important in the final phase of melting process.

ELECTROMAGNETIC FIELD

In order to calculate the thermal part of inductive heating in the load, Joule heat sources must be known. Moreover, inductive heating creates the Lorentz force influencing the convective transport. For these reasons, the distribution of electromagnetic field should be calculated. Joule heat is given by the distribution of electromagnetic field. The common way in axial symmetric case is to use vector potential $\mathbf{A}(t) = \mathbf{A}e^{i\omega t}$ with only one vector component, which in non-magnetic material gives following equation

$$\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial (rA)}{\partial r}\right) + \frac{\partial^2 A}{\partial z^2} = \mu_0 \sigma(T)\left(i\omega A + \frac{1}{r}\frac{\partial U}{\partial \varphi}\right), \tag{1}$$

where A is azimuthal component of complex vector potential $\mathbf{A}$; U is the scalar potential the azimuthal gradient of which differs from zero only in the inductor. As the inductor consists from one massive conductor, the distribution of current density in the inductor must be calculated additionally using the independence of the scalar potential in the cross-section of it. The normalized vector potential $a = rA$ decreases far from axis of symmetry very slowly as $1/r$. Henceforth, the homogeneous boundary condition of third kind is used instead at infinite surfaces assuming the system as magnetic dipole. The conductivity of the oxide material rapidly increases with temperature, which requires frequent recalculation of electromagnetic field during heating of the load. The conductivity of the most of oxides and glasses increases exponentially with temperature like semiconductors. At very high temperatures, the conductivity saturates. The conductivity depends very much on the impurities and microscopic modifications of the oxide material. Therefore, the exact knowledge of the properties of oxide material remains a serious difficulty.

HEAT TRANSFER

The equation equation of thermal equilibrium including Joule heat sources and convective transport in axis symmetrical case is

$$\rho c_p\left(\frac{\partial T}{\partial t} + v_r\frac{\partial T}{\partial r} + v_z\frac{\partial T}{\partial z}\right) = \frac{1}{r}\frac{\partial}{\partial r}\left(r\lambda(T)\frac{\partial T}{\partial r}\right) + \frac{\partial}{\partial z}\left(\lambda(T)\frac{\partial T}{\partial z}\right) + \frac{\sigma(T)|A|^2\omega^2}{2}. \tag{2}$$

The thermal conductivity usually essentially depends both on temperature and porosity. The first dependence is especially important for glass, where radiation heat transfer becomes dominant at temperatures above 600 K. Radiation heat losses from the top of melt are included while the surface temperature can reach 2000 K yielding considerable amount of heat losses. Despite the other molten oxides could be transparent, too, the local or non-local radiation heat transfer inside the load this case is neglected due to lack of data. Additional gas burner with constant power and Gaussian spatial distribution can be included. Phase transformations create additional numerical difficulties in calculations.

SINTERATION

The initial material is usually like a powder with some porosity coefficient $\Pi \in [0,1]$ given by volume fraction of void. The porosity decreases as the temperature of the melt increases. Because the exact behaviour of sinteration below melting point is not known, it is assumed that irreversible decrease of porosity occurs only at melting point. Melt filtration is not included, which however could influence the process especially at burn-out of Mo ring. Different model for change of porosity is used for glass melting, where the distinct phase transition is not present. The decrease of porosity for amorphous glass grains is set proportional to the pressure P and inversely proportional to the dynamic viscosity μ:

$$\frac{\partial \Pi}{\partial t} \sim -\frac{P}{\mu(T)}. \tag{3}$$

The additional multiplier with an order of 1 depends in complicated way on geometric factors. The viscosity exponentially decreases with temperature for the amorphous substance like glass. Therefore, the considerable change of porosity (sinteration) occurs only at temperatures above 1000 °C for glasses, while the sinteration at lower temperatures lasts for years. In principle, Eq. 3 can be generalised also for the oxides like ZrO_2, where the viscosity of condensed material is much higher than the viscosity of the melt, but it decreases with temperature, too.

Discrete addition of new material is considered both in experimenthal furnace and the simulations due to notable (~30 %) shrinkage of initial porous material. The added porous and cold material reduces radiation losses and keeps the load more close to the centre of the inductor. Thus, it improves the energy balance of the inductive heating.

MELT FLOW

In order to account for the convective transport in Eq. 2, the flow of the melt should be calculated which is created by the body forces. The current induced by magnetic induction leads to the appearance of Lorentz force in melt material. Because of high frequency melting we take a time average values of these quantities. In addition to the Lorentz force, thermo-gravitational force is present especially for melt with relatively high linear expansion coefficient α. Thus the total resulting force density in Bouyoancy approach is

$$\vec{f} = \vec{j} \times (\nabla \times \vec{A}) - \vec{g}\rho_0 \alpha (T - T_0), \tag{4}$$

where g is the gravitational acceleration, ρ_0 – density at reference temperature of melt T_0. Resulting force creates melt motion inside the crucible. The flow is only considered as transient, but there is no significant differnce with stationary flow since the heating up of melt by inductive melting occurs slowly in an order of an hour. The two velocity components and pressure follow from momentum equation

$$\rho\left(\frac{\partial\vec{v}}{\partial t}+(\vec{v}\nabla)\vec{v}\right)=\nabla(\mu(T)\nabla\vec{v})-\nabla p+\vec{f} \quad (5)$$

and the continuity equation

$$\nabla\vec{v}=0\,. \quad (6)$$

The boundary conditions of velocity are chosen in correspondence with physical model, i.e., non-slip boundary conditions along solid boundaries and gradient free boundary condition for tangential velocity on axis of symmetry and top of the melt. Either a variant of SIMPLER method [3] or direct solution of both Eq. 5-6 is chosen. In the first case the difficulties are situation-dependent convergence to divergence free flow and prpoer choice of pressure boundary conditions. In the second case, the major difficulty is how to avoid the slight singularity of the equation system. Here, a small infinitasemal lapplacian of the pressure $\varepsilon\Delta p$ is added to continuity equation Eq. 6 that avoids the singularity. The flow usually is present only in the part of the load especially at the beginning of the melting process. Therefore, the flow equations are calculated only in the area with lowest viscosity.

It was found that moderate inductive heating is necessary in the initial phase to obtain the temperature distribution as homogeneous as possible before pure electromagnetic heating can start [1]. Different heating methods are considered in next two sections.

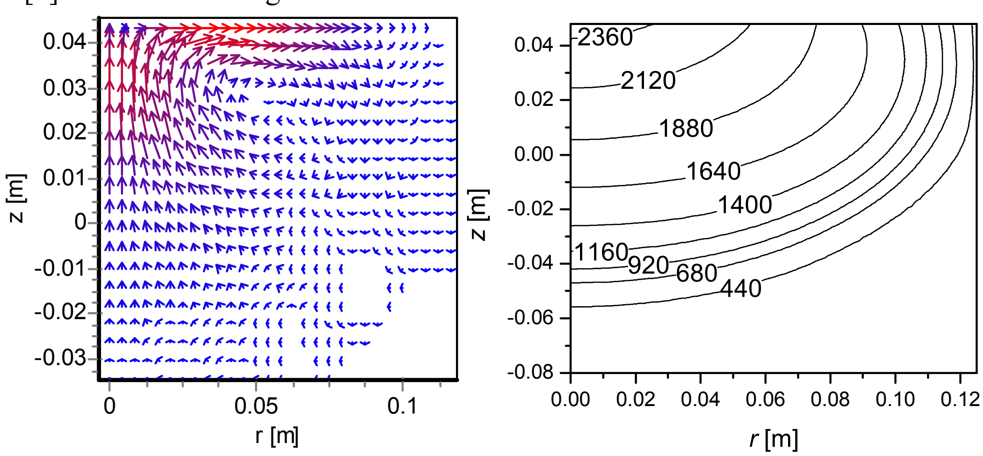


Figure 2. Flow patterns at the initial stage of glass melting with corresponding temperature distributions. Initial heating by gas burner accompanied with inductive heating. Only significant part of the furnace is shown in flow patterns.

INITIAL HEATING BY GAS BURNER

As mentioned in the introduction initial pre-heating of the load before pure inductive heating can start can be performed by the gas burner. The gas burner can be placed near the top of the melt as shown in Figure 1. Certainly, the highest amount of heat in the load is produced directly below the gas burner and its effectiveness reduces by the distance from the centre. It is assumed in simulations that gas burner has Gaussian spatial distribution of heating power. Gas burner heats only the top surface of the load if non-local heat transfer by radiation is neglected. However, the pure electromagnetic heating can start more easily if the heating occurs ins ide the average vertical position of the inductor close to its surface. Therefore, quite slow heating by gas burner is preffered that will enable to increase the temperature also in the centre of load by conduction. If the conduction of the load is very small as for oxides like ZrO_2, a large amount of heat will be already lost before pure inductive heating. The heating by gas burner is more effective if the load conductivity rapidly increases with temperature as for glasses, where radiation heat transfer becomes dominant for temperatures above 600 K. It is possible to use the inductive heating simulataneously with gas burner. Despite the inductive heating of cold load is ineffective, it will slightly extend the heated area by gas burner towards the inductor as shown in Figure 2. Then the pure inductive heating after removing of gas burner can start much more easily. Moreover, the use of inductive heating during the heating by gas burner enables to estimate the condition of the load by monitoring of the inductive power at constant voltage or current. If the inductive power starts to increase rapidly overcoming certain threshold, one can be sure that the load is well pre-heated and gas burner can be swithced off. The example of the increasement of power and other parameters is shown in Figure 3.

Figure 2 shows the characteristic flow pattern during heating by gas burner. Flow is present only in the hotter part of the load where the viscosity coefficient is much lower. Thermograviational force leads to melt motion from the centre at the top of the load. The flow direction would be opposite if the heating occurs at the middle of the load as in the final phase of the process. Only one or two vortexes of the flow are present since viscous kind of glass is considered. The vortex directions depend on whether electromagnetic or thermo-gravitational force is dominating. Linear expansion coefficient of considered glass was 10^{-5} 1/K, which lead to dominance of thermo-gravitational force over electromagnetic one.

The downward peaks in maximal and surface temperature, and power in Figure 3 occurs at the time moments when new cold material is put over the load. The adding of new material lessens the radiation losses and improves the total energy balance.

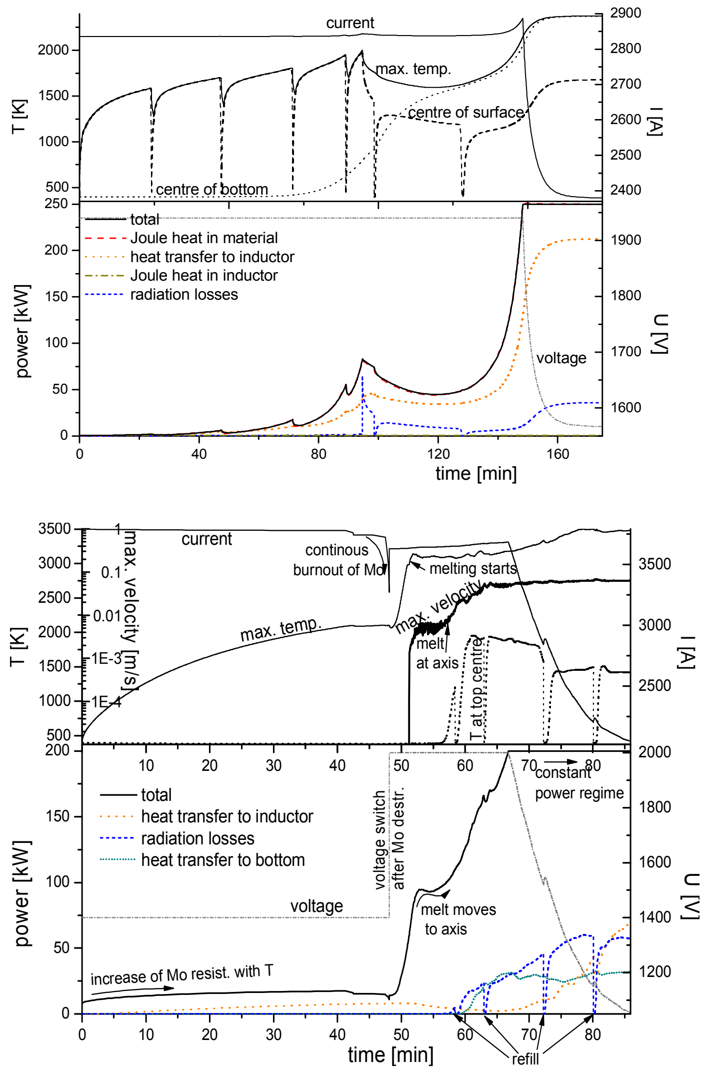


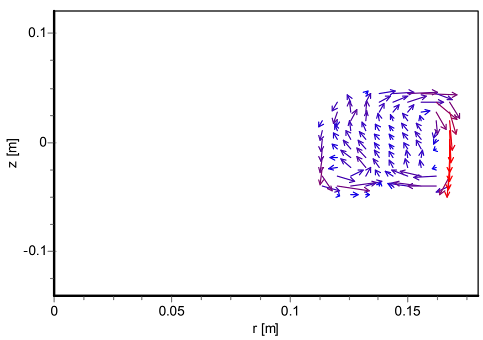
Figure 3. Behavior of power parameters and temperatures during inductive heating of glass with initial heating by gas burner (left) and in ZrO_2 (right) with initial heating

INITIAL HEATING BY METAL RING

The initial heating by gas

burner is innefictive of oxides like ZrO_2 with low thermal conductivity also at high temperatures. In this case it is much better to heat the bulk of the load directly, while thermal transfer to the othe rareas is slow. On of the possibilities is to put a conductive material, e.g, metal in to the load. The Joule heat produced in the conductive material heats up the surrounding oxide close to the temperatures when inductive heating of oxide itself can start. The metalic conductor however spoils the further inductive heating and processing of the required oxide. Therefore, it is preferable that metalic material losses its abbility of conduction when sufficiently high temperature is already present in to oxide load. One of such possibilities is to use Mo conductor inside the ZrO_2 or $ZrSiO_4$ load, which oxidises in this environment at high temperatures. Afterwards, we can neglect the presence of Mo conductor. The most appropriate shape of the metal conductor is ring placed axis symmetric into the inductor. It absorbs the electromagnetic energy more effectively in this case.

We have noted in paper [1] that it is necessary to get the temperature distribution as homogeneous as possible during initial heating, in order to decrease the maximal voltage or current for continuing the heating process. In opposite case the load could cool down after the initial heating is finished, because the material has significant conductivity only at some small region. Therefore, it is necessary to use moderate initial heating of metal ring.



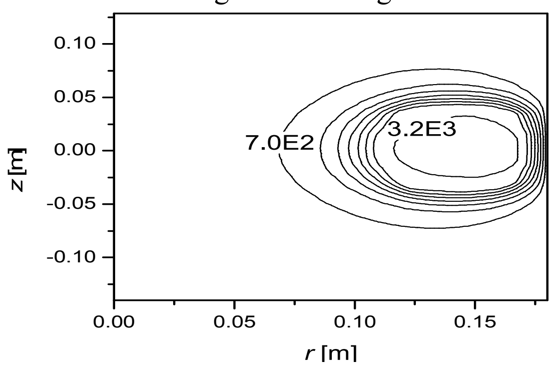


Figure 4. Flow pattern in melting of ZrO_2 at the time moment when melting is just started with corresponding temperature distribution (α=10^{-6} 1/K, μ=10^{-2} kg/(m·s)). The load is preheated by Mo ring.

The viscosity coefficient of ZrO_2 is very high below melting point and almost constant above melting point. Therefore, notable velocity is only in the molten region of ZrO_2 load. Its size increases but flow pattern significantly depend on linear expansion coefficient α and dynamic viscosity coefficient μ. The properties of molten ZrO_2 are poorly known. Hence, we can study only on the influence of these parameters [3]. The melting starts, where the temperature is the highest, i.e., where the ring is burned out. Next, the molten area moves either towards the centre if thermal expansion of the load is small or upward in opposite case. It must be noted that filtration of melt in porous material is neglected as the condensed part of melt could quickly fill the pores near the fluid front. If the melt moves to the centre, the resulting power decreases at constant voltage or current of inductor as the electromagnetic coupling becomes weaker. Afterwards, the area of molten region becomes wider until the stationary distribution at constant power is reached. The constant voltage regime is changed to constant power regime at the moment when the total power of the inductor exceeds certain value.

The example of inductive heating of ZrO_2 in the furnace is shown in Figure 4. The maximal amplitude of voltage is chosen 2000 V. The maximal current in this simulation does not exceed 3800 A. At critical stage, the active power of the inductor rapidly starts to grow. Therefore, when the power reaches 200 kW, constant voltage is changed by this maximal power. Figure 4 shows also the behaviour of maximal temperature in the load. When the area of molten region is quite small, the maximal temperature reaches 3500 K when the flow intensity of melt is relatively low.

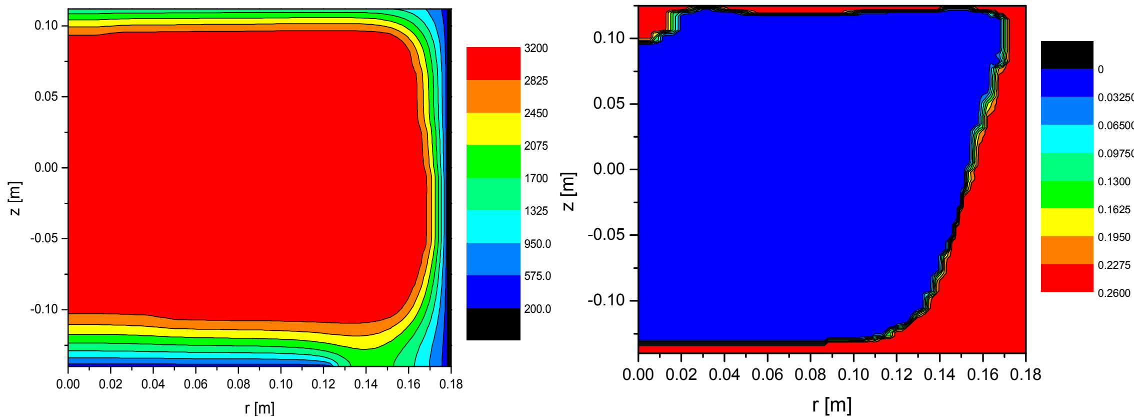


Figure 5. Characteristic stationary temperature (in K) distribution and shape of skull layer (red), i.e, layer with nonzero porosity. The skull layer at the bottom of inductor is thicker because of specific flow pattern and weaker electromagnetic field in load

STATIONARY CASE AND SKULL LAYER

The situation in the stationary state is little dependent how the load has been initially heated. Therefore, the stationary distribution should be similar for initial heating with gas burner or metal ring. The analysis of stationary distribution is quite important for study of the skull layer, which in other hand significantly depend on boundary conditions, melt flow intensity and direction, power of the inductor, melt properties. The characteristic temperature distribution, skull layer, and flow patterns are shown in Figures 5-6. The skull layer at the bottom of inductor is thicker because of specific flow pattern and weaker electromagnetic field in lower part of the load. The thickness of the skull layer varies from a millimiter scale to a centimetre.

Figure 3 show that in the final phase heat transfer losses to inductor, cooled bottom, and radiation losses vary from 20 to 80 kW each at total power of about 200 kW for about 20-40 l load. Joule heat losses in copper inductor ~1-2 kW is negligible constituent.

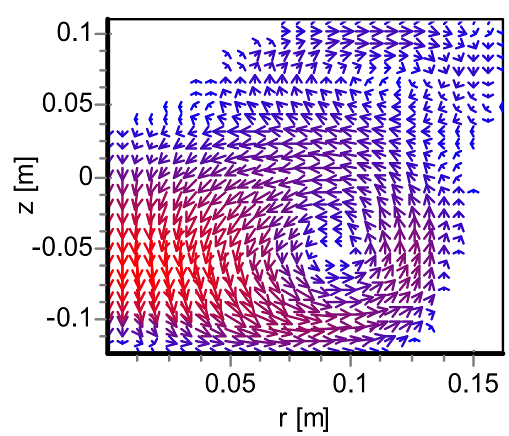


Figure 6. Characteristic velocity distribution in load with dominant electromagnetic force

OPTIMISATION

The numerical simulation allows optimisation of furnace parameters [3]. As example Figure 7 – left shows the optimisation of frequency. Each curve represents the corresponding average temperature of the melt. The largest average temperature is close to processing temperature of the melt. The used fraction of energy means ratio between total increase of internal energy of melt with respect to total amount of energy supplied both by the inductor and gas burner. The optimal frequency for considered system, if other parameters are fixed, appears to be 300-400 kHz. Higher frequency is more efficient for heating in the starting phase when the conductivity of the load is quite low. The irregularities in these curves are caused by discrete addition of new material. Figure 7 – right shows the optimisation in case of initial melting by Mo ring. The optimisation parameter is initial amplitude of voltage. If the initial voltage does not fall in the shown range, the melting does not start at given maximal voltage of the inductor. We can see that maximum efficency is close to the highest voltage in the possible diaposon of voltages. Other possible parameters for optimisation include geometry of the inductor, filling fraction with load in the inductor, voltage-current regime, Mo ring position and size, etc. However, due to technical restrictions of the furnace (limited range of voltage, current, frequency, sizes, etc.) constrained optimisation is required.

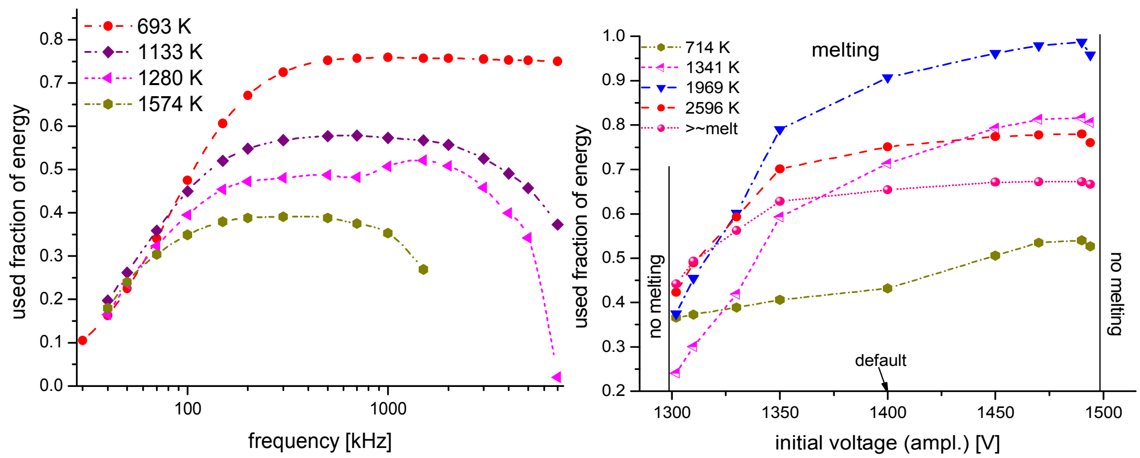


Figure 7. Left - optimisation of furnace parameters: frequency. The curves correspond to the given average temperature of the load. Right - optimisation of initial voltage in presence of Mo ring at constant other parameters. The voltage outside the plotted range does not lead to melting. The curves denote heating up to the given average temperature or certain fraction of melt as for last curve.

CONCLUSION

Basing on the axialsymmteric model of inductive heating in inductor crucible, the influence of different regimes of the processes have been studied. The model is based on simultaneous calculation of electromagnetic, temparature, and flow fields of incompressible melt. The simulations showed high importance of the pre-heating regime of the load, and it should be perfomed with moderate power both in heating by gas burner and heating by metal ring. The sinteration of the porous material and addition of new material has been included, which, generally, improves the energy balance. The numerical simulations enable to optimise various furnace parameters and regime of heating.

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