

Thermal Conductivities of Slags

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ABSTRACT

The thermal conductivity data for solid and molten slags have been collated and evaluated. The various factors affecting the measurement of thermal conductivities are discussed.

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SYMBOLS, ABBREVIATIONS

a	thermal diffusivity	m^2s^{-1}
C_p	heat capacity	$\text{JK}^{-1}\text{kg}^{-1}$
d	thickness of sample	m
k	thermal conductivity	$\text{Wm}^{-1}\text{K}^{-1}$
k_{eff}	effective thermal conductivity	
k_L	lattice (phonon) thermal conductivity	
k_m	thermal conductivity of liquid at melting point	
k_R	radiation (photon) conductivity	
k_{el}	electronic thermal conductivity	
L	mean free path of phonons	m
n	refractive index	
Q	heat flux density	Jm^{-2}
T	temperature	K, °C
T_g	glass transition temperature	
$T_{\text{liq}} \downarrow$	Liquidus temperature, denoted by $\downarrow$ in figures	
v	average velocity of sound in the melt	ms^{-1}
x	mole fraction	
α	absorption (or extinction) coefficient	m^{-1}
ρ	density	kg m^{-3}
ESR	Electro Slag Remelting	
LS	Line Source Method	
LP	Laser Pulse Method	
MB	Modulated Beam Method	
RHF	Radial Heat Flow Method	
RW	Radial Wave Method	
sc	Supercooled Melt	
%	Mass Percent	

1 INTRODUCTION

It is customary to measure thermal conductivities (k) from measurements of either heat flux (Q) or thermal diffusivity (a) using the relationships given in Equations (1) and (2), respectively.

$$Q = -k_{\text{eff}} (dT/dx) = \frac{-k_{\text{eff}} \cdot \Delta T}{d} \quad (1)$$

where k_{eff} is the effective thermal conductivity, ΔT the temperature gradient over the distance, d .

$$k_{\text{eff}} = a_{\text{eff}} C_p \cdot \rho$$

where C_p and ρ are the heat capacity and density, respectively. The term 'effective' is used since the thermal conductivity and diffusivity can contain contributions from various heat transfer mechanisms, viz, the lattice ('phonon') thermal conductivity, k_L , the radiation ('photon') conductivity, k_R , and the electronic thermal conductivity, k_{el} . Under 'optically thick' conditions k_{eff} can be derived (1,2) from Equation (3).

$$k_{\text{eff}} = k_L + k_R + k_{el}$$

1.1 THERMAL CONDUCTION MECHANISMS

1.1.1 Lattice or 'phonon' thermal conductivity (k_L)

Heat is transferred through a medium by phonons, which are quanta of energy associated with each mode of vibration in the sample. This mode of conduction is thus referred to as thermal, phonon or lattice conduction. Scattering of the phonons causes a decrease in the thermal conductivity which is therefore sensitive to the structure of the sample. Scattering of the phonons can occur by collisions of the phonons with one another, or by impact with grain boundaries or crystal imperfections, such as pores. Thus a low-density, highly-porous material will have a low thermal conductivity. In glassy, non-crystalline materials it has been suggested (3) that thermal conductivity decreases as the disordering of the silicate network increases.

1.1.2 Radiation conductivity (k_R)

Measurements of the thermal conductivity of glasses were found to be dependent upon the thickness of the specimens used, (Figure 1). This behaviour was ascribed to the contribution from radiation conductivity, k_R , which can occur in semi-transparent media like slags. Radiation conductivity occurs by a mechanism involving absorption and emittance of radiant energy by various sections through the medium (1,2). Consider a thin section in the slag, radiant energy absorbed by the section will cause it to increase in temperature and consequently radiant heat will be emitted to cooler sections. This process can occur continuously through the medium and it is obvious that the energy transferred in this way will increase with increasing number of sections (ie increasing thickness) until the point is reached where k_R attains a constant value. At this point the slag is said to be "optically

thick", and this is usually considered to occur when $\alpha d > 3.5$, where α and d are the absorption coefficient and thickness of the slag sample, respectively.

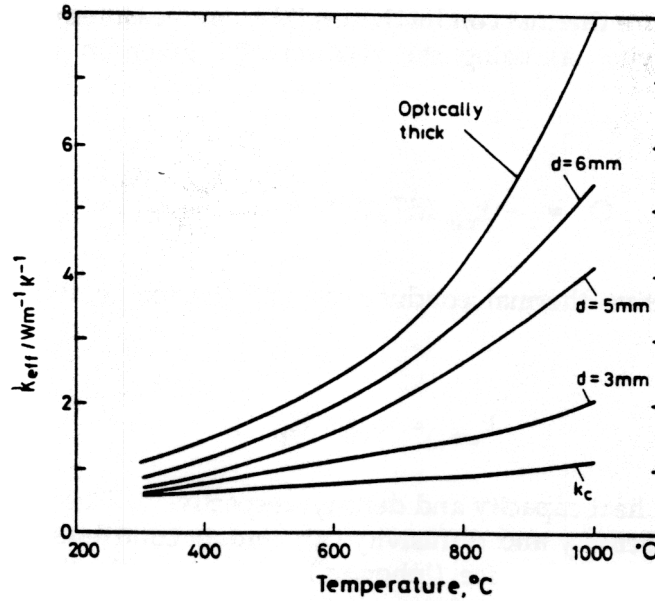


Figure 1 - The dependence of the effective thermal conductivity upon the thickness of the specimen

At high temperature, radiation conduction is frequently the dominant mode of heat transfer, eg in glassmaking more than 90% of the thermal transfer occurs by radiation conduction. The radiation conductivity can be calculated for an optically-thick sample if steady-state conditions apply and if it is assumed that the absorption coefficient of the medium, α , is independent of wavelength, λ , ie grey-body conditions obtain. For these conditions k_R can be calculated by use of Equation (4), where σ and n are the Stefan-Boltzmann constant and refractive index, respectively.

$$k_R = \frac{16 \sigma n^2 T^3}{3 \alpha} \quad (4)$$

The above equation can not be applied to 'optically thin' conditions (ie $\alpha d < 3.5$); for these conditions, Equations (5) and (6) may be used (4,5) in conjunction with tabulated data for the reflection coefficient, $R (= 1 - \epsilon)$ of the boundary and where ϵ is the emissivity and τ is the optical density

$$k_R = 8n^2\sigma T^3 \frac{(1-R)}{2(1+R)} - \frac{\tau(1-4R + R^2)}{(1 + R)^2} \quad (5)$$

$$k_R = \frac{16n^2 \sigma T^3}{3} \gamma(\epsilon, \tau) \quad (6)$$

Electronic conductivity (k_{el})

It has been suggested that slags containing high concentrations of Fe^{2+} ions could behave like semiconductors in that heat could be transferred by conduction electrons, holes, etc. However, on the basis of electrical conductivity data (6) it can be concluded that electronic conduction only becomes significant when the FeO content of the slag is $> 70\%$.

2 EXPERIMENTAL METHODS (7,8,9)

The techniques for the determination of thermal conductivity can be divided into three classes: (i) steady-state methods, (ii) non-steady state methods, and (iii) transient methods. The steady-state methods usually yield k_{eff} values and the non-steady state techniques usually produce thermal diffusivity (a_{eff}) values, which can be converted to thermal conductivity values by use of Equation 2.

2.1 STEADY STATE TECHNIQUES

Linear heat flow method

In this method two disc-shaped specimens are placed on either side of an electrically-heated plate and the temperature profiles across the samples are monitored by thermocouples sited on both faces of the specimen. The apparatus is well insulated to minimise heat losses. In some versions of this method, the total heat flux passing through the samples is determined by calorimeters in contact with the specimens. When high-temperature measurements are required, this technique is usually operated as a comparative method (10).

Radial heat flow (RHF) method

In this method the specimen is in the shape of a hollow cylinder (Figure 2) which is positioned in the annulus between two coaxial cylinders with the internal cylinder acting as a radial heat source (11,12). The temperature profile across the specimen is determined by thermocouples placed on the inside walls of the two cylinders. This method requires a large isothermal zone in the furnace, which is difficult to achieve at high temperatures. The results contain contributions from radiation conduction and when this technique is used for measurements on liquids, errors can occur from convective heat transfer.

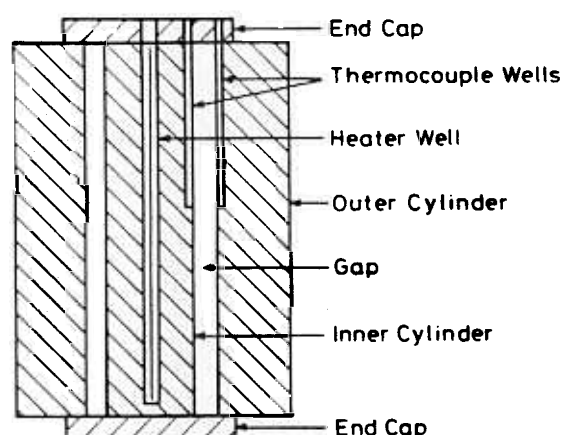


Figure 2 - Schematic representation of radial heat flow (or concentric cylinder) apparatus

2.2 NON-STEADY STATE METHODS

Radial wave (RW) method

In this method the slag is placed in a cylindrical crucible situated in the isothermal zone of a furnace, and thermocouples are located on the walls and along the geometric axis of the crucible (the slag). The outside wall of the crucible is then subjected to a variation of temperature and the variation in temperature of the central thermocouple is monitored. There is a phase shift between the input and output which is related to the thermal diffusivity of the slag. In the modification of this apparatus used by Elliott and co-workers (13,14) the periodic variation in temperature is produced in a wire running along the central axis of the cylindrical crucible, and the phase shift is measured by thermocouples located on the walls of the crucible. The thermal diffusivity values obtained with this method may be vulnerable to errors arising from convective and radiative heat transfer.

Modulated beam technique

In this method the specimen is in the form of a disc, which is maintained at a constant temperature, whilst the front face is subjected to a laser beam which produces a periodic variation in temperature of constant frequency. The phase shift between this input and the signal from a temperature sensor in contact with the back face is determined. By carrying out measurements at two or more frequencies, Schatz and Simmons (15) were able to derive values of both a_{eff} and the extinction coefficient. If this method were applied to measurements for liquids, it too would be prone to errors caused by convection.

2.3 TRANSIENT METHODS

In these experiments the transient of the temperature-time curve produced by an input of energy is monitored during the short duration of the experiment (10 seconds or less).

Laser pulse (LP) method

The sample used in this technique is usually a disc of 10-15 mm diam $\times$ 1-2 mm (Figure 3a) which may be coated with metallic coatings on both planar surfaces (16,17). A laser pulse is directed onto the front face of the specimen and the temperature of the back face is monitored continuously. The temperature transient (Figure 3b) exhibits a maximum (ΔT_{max}) and the thermal diffusivity (a) can be calculated from Equation (7) where $t_{0.5}$ is the time to reach $0.5 \Delta T_{\text{max}}$ and d is thickness of the specimen. Corrections are usually made to account for radiation losses.

$$a_{\text{eff}} = \frac{1.37 d^2}{\pi^2 t_{0.5}} \quad (7)$$

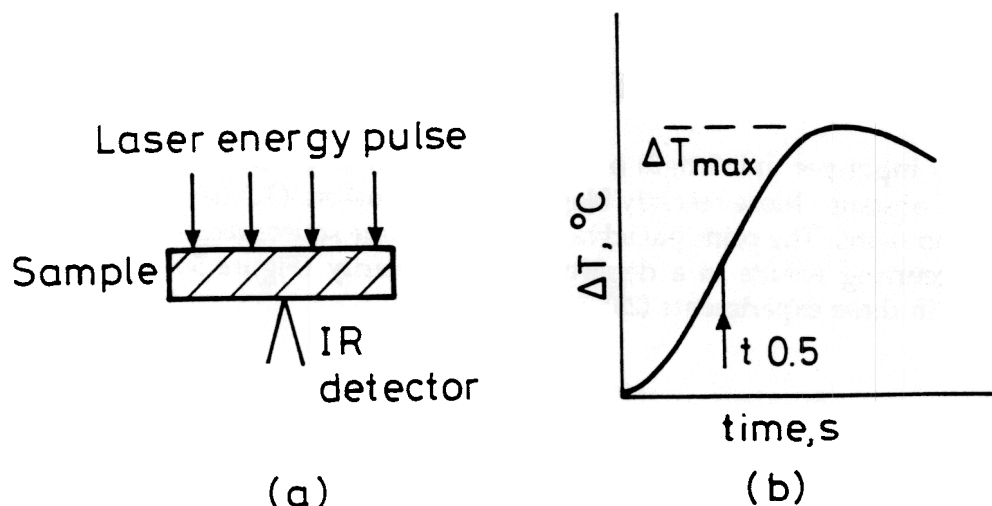


Figure 3 - The laser pulse (LP) method used for determination of the thermal diffusivities of solids showing (a) the specimen assembly and (b) the temperature transient

For measurements on liquid slags (18,19) the laser pulse of short duration is directed onto platinum disc on the surface of the slag (Figure 4a) and the temperature transient of this disc is monitored continuously (Figure 4b). Numerical analysis have been used to separate k_R and k_L contributions in these experiments (18,19).

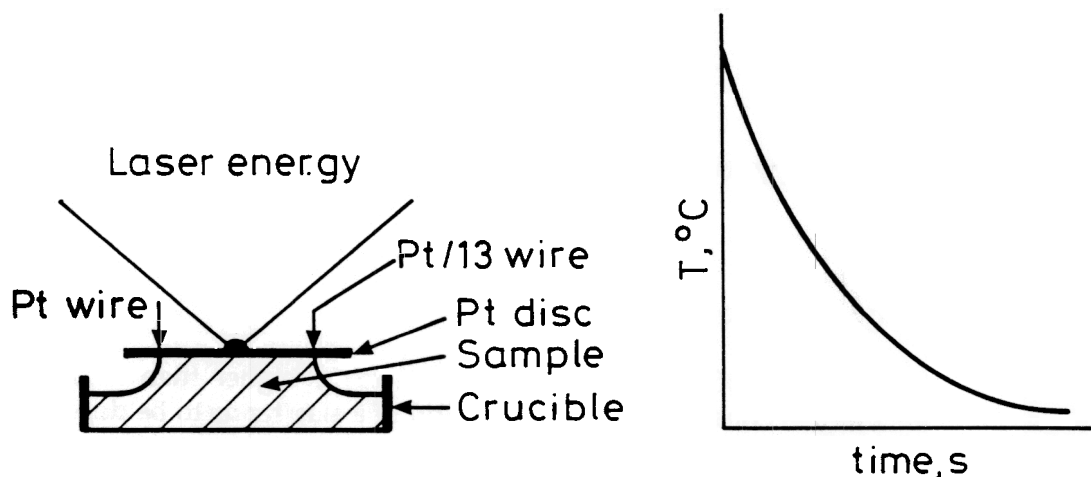


Figure 4 - The laser pulse method used for determinations of the thermal diffusivities of liquids showing (a) the cell assembly used and (b) the temperature transient

2.3.2 Line source (LS) method

In this method (20,21) current is applied for a short period (cals) to a long thin wire which is immersed in a cylinder of molten or solid slag (Figure 5a). The wire acts as both heating element and resistance thermometer and the temperature of the wire is monitored continuously during the application of current. The thermal conductivity can be calculated from Equation (8) but in practice it is usually derived from the slope ($\cong 1/k$) of the plot of ΔT as a function of $\ln$ time (Figure 5b).

$$\Delta T = \frac{q}{4 \pi k} \ln \frac{4at}{r^2 C} \quad (8)$$

where q is the heat input per unit length of wire, r is the radius of the wire and $C = \exp.\gamma$ where γ is Eulers Constant. More recently Nagata and co-workers (22) have used a thin Pt strip in place of a hot wire. The principal advantages of the line source technique are (i) that any convection occurring results in a departure from linearity (Figure 5b) and (ii) k_R is thought to be low in these experiments (23).

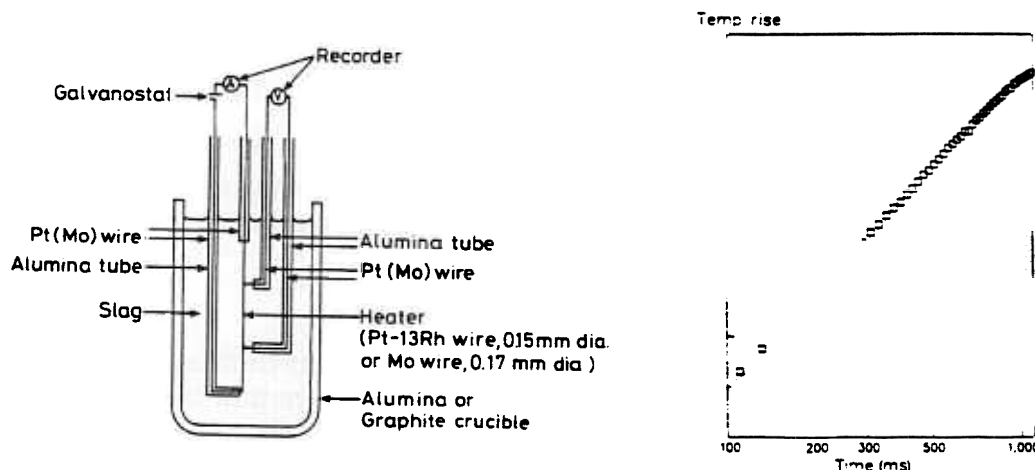


Figure 5 - The line source method (a) schematic drawing of the probe (21) and (b) temperature transient

3 FACTORS AFFECTING THERMAL CONDUCTIVITY

3.1 SOLIDS

Crystallinity of sample

The thermal conductivities of crystalline phases tend to be higher than those for glassy phases. Thus the thermal conductivity (or diffusivity) of a slag tends to be dependent upon its thermal history.

Porosity of sample

The thermal conductivities of crystalline materials are reduced by the presence of flaws in the lattice such as pores, microcracks. Several equations have been proposed for calculating the effective thermal conductivities, of sintered and powder samples from the thermal conductivity of the theoretically dense solid and the porosity of the sample (24,25).

Glass transition temperature (T_g)

It can be seen from Figure 6 that the thermal conductivity of glassy slags goes through a maximum at the glass transition temperature, T_g which is associated with the large increase in thermal expansion coefficient (ie "free volume") which occurs above T_g (26).

LIQUIDS

Degree of polymerisation of the sample

The thermal conductivity of a liquid at the liquidus temperature, k_m , increases with increasing SiO_2 content, ie increasing degree of polymerisation (26). The (NBO/T), ie the number of non-bridging oxygens per tetrahedrally-co-ordinated atom (eg Si, Al) is a measure of the degree of depolymerisation. It can be seen from Figure 6 that k_m decreases as (NBO/T) increases.

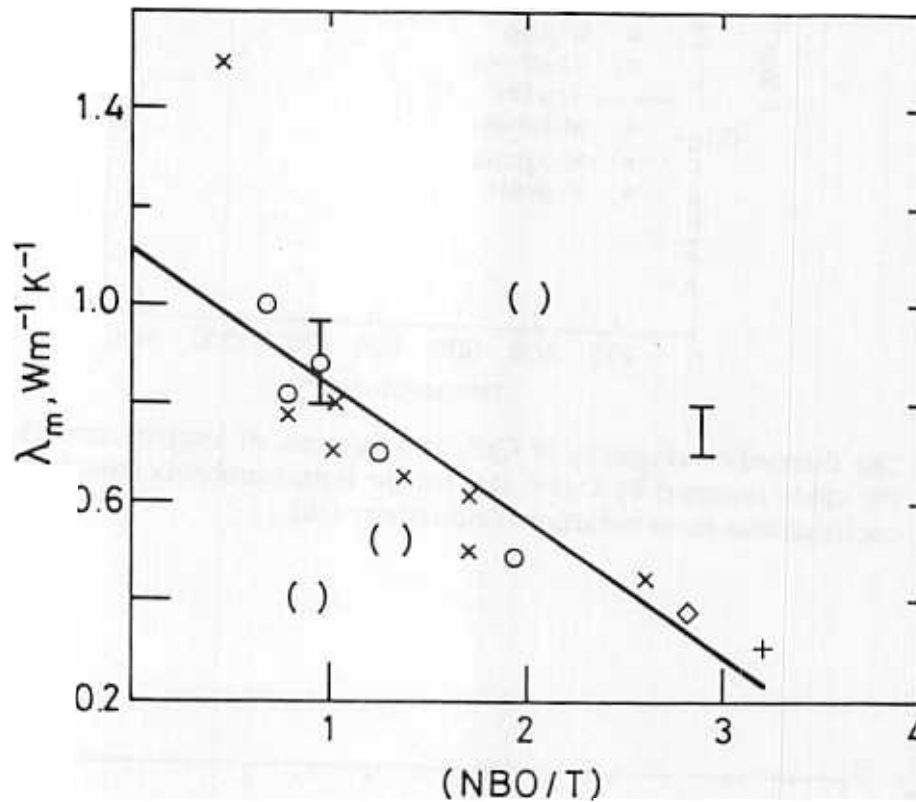


Figure 6 - The thermal conductivity at the liquidus temperature, k_m as function of (NBO/T) ratio, ie the number of non-bridging oxygens per tetrahedrally-co-ordinated atom

3.2.2 Relation to velocity of sound

The relationship between the velocity of sound (phonons) in the slag (v) and the thermal conductivity (k_L) is given in Equation (9) where L is the mean free path of the phonons. Values of L derived from k_m values and velocity of sound data were found to approximate to the Si-O interatomic distance (26).

$$k_L = \frac{1}{3} (C_p \cdot v \cdot L) \quad (9)$$

4 THERMAL CONDUCTIVITY DATA

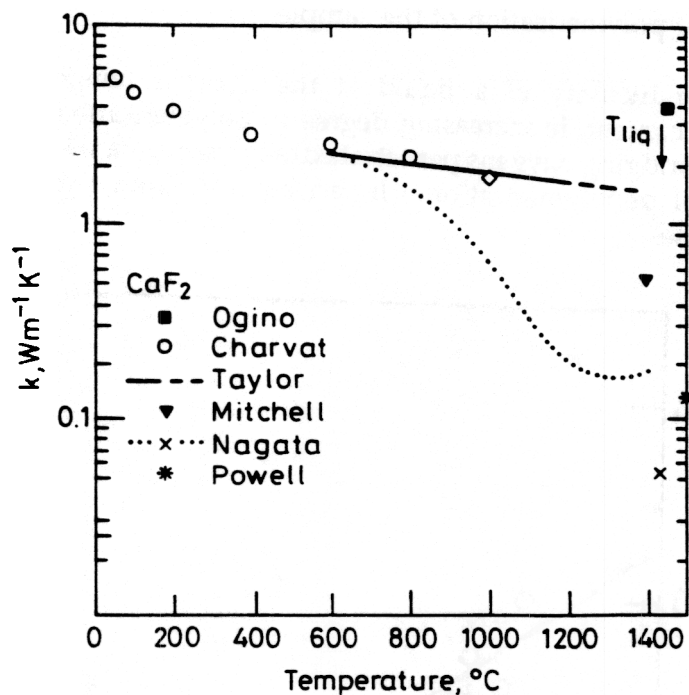


Figure 7 The thermal conductivity of CaF_2 as a function of temperature (11,17, 27-30) the value reported by Ogino (11) for the liquid probably contains significant contributions from radiation conductivity (30).

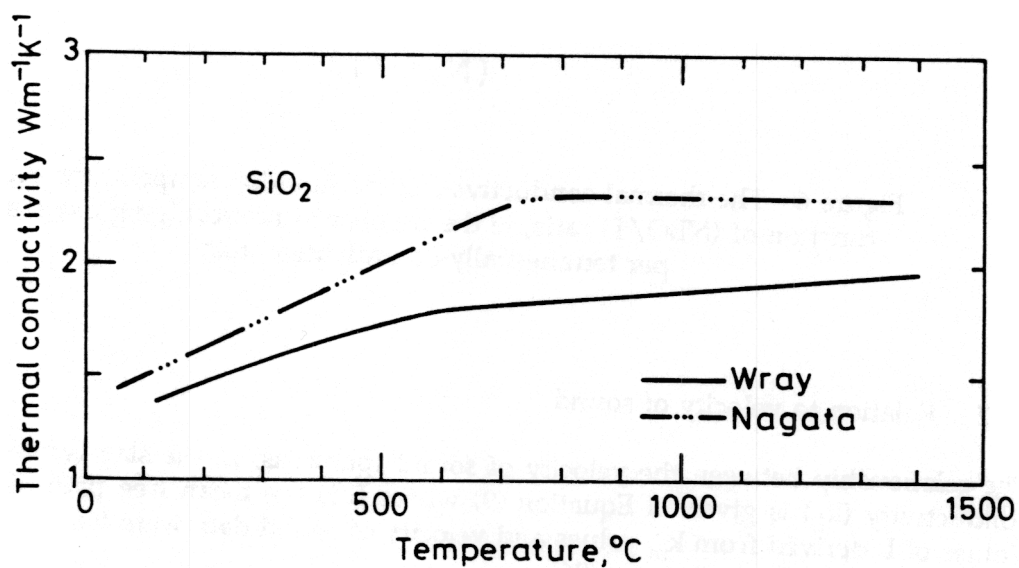


Figure 8 Thermal conductivity of SiO_2 as a function of temperature (20,31).

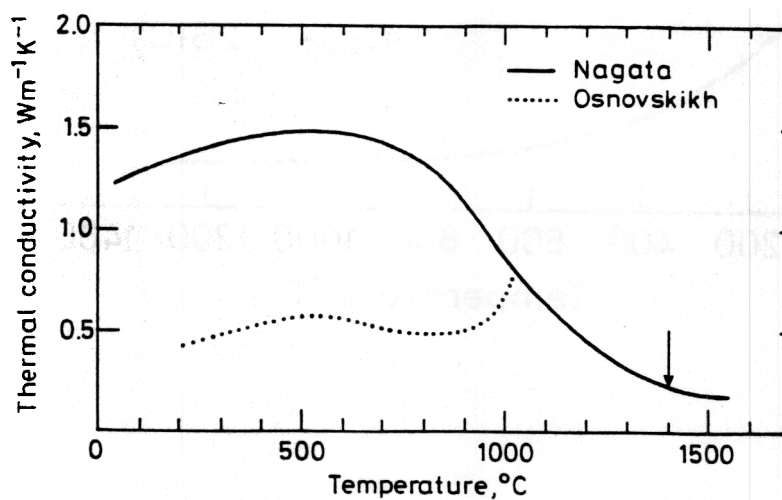
$\text{Al}_2\text{O}_3\text{-CaO}$ 

Figure 9 Thermal conductivity of slags containing 50% Al_2O_3 - 50% CaO as a function of temperature (20,32).

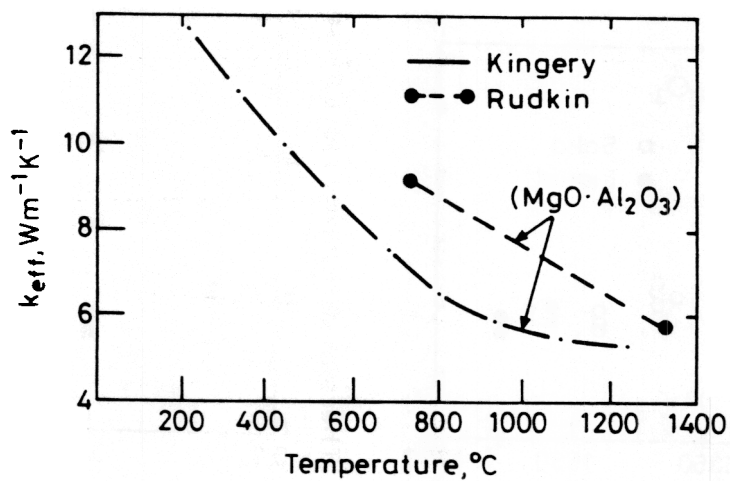
 $\text{Al}_2\text{O}_3\text{-MgO}$ 

Figure 10 Thermal conductivity as a function of temperature for spinel ($\text{MgO} \cdot \text{Al}_2\text{O}_3$) (10), and values derived (30) from thermal diffusivity measurements (33).

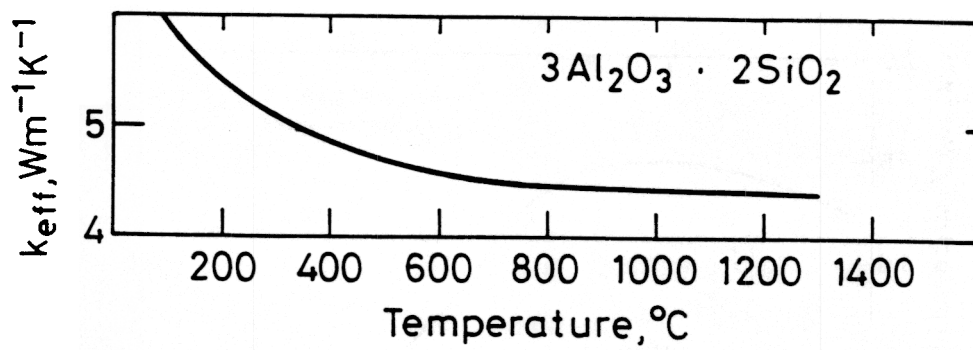
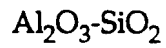


Figure 11 Thermal conductivity of sintered mullite ($3\text{Al}_2\text{O}_3\cdot\text{SiO}_2$) as a function of temperature (10).

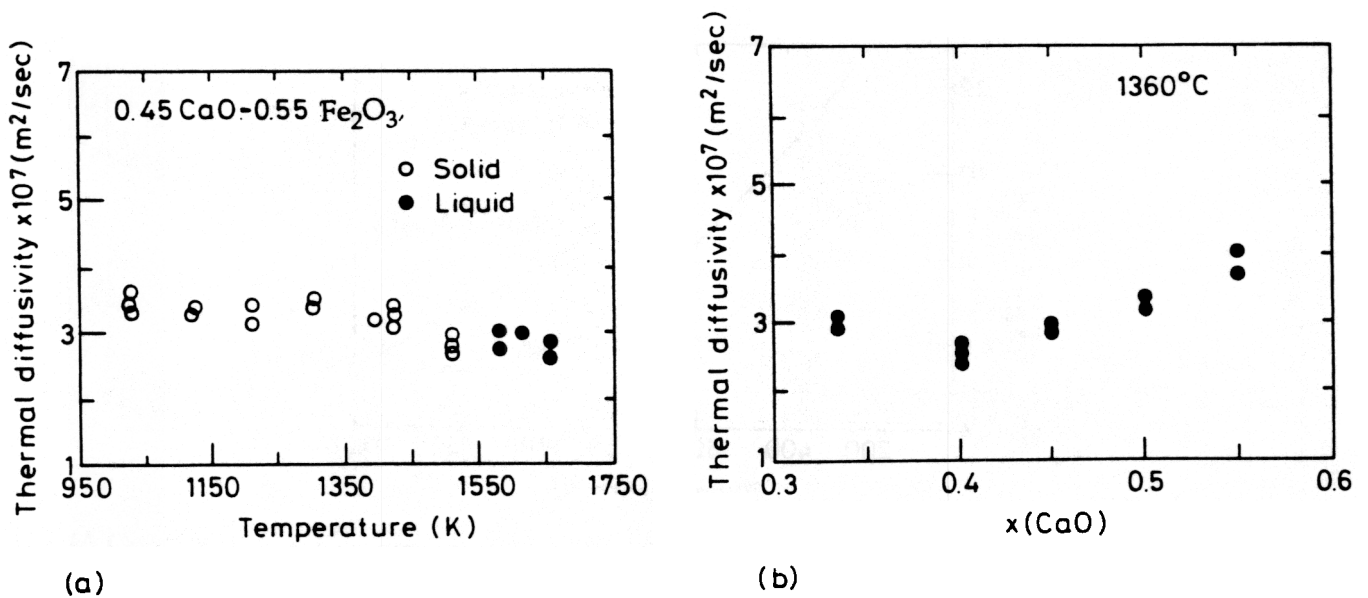
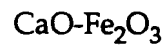


Figure 12 Thermal diffusivity of $\text{CaO-Fe}_2\text{O}_3$ slags as a function of (a) temperature (b) CaO content (34).

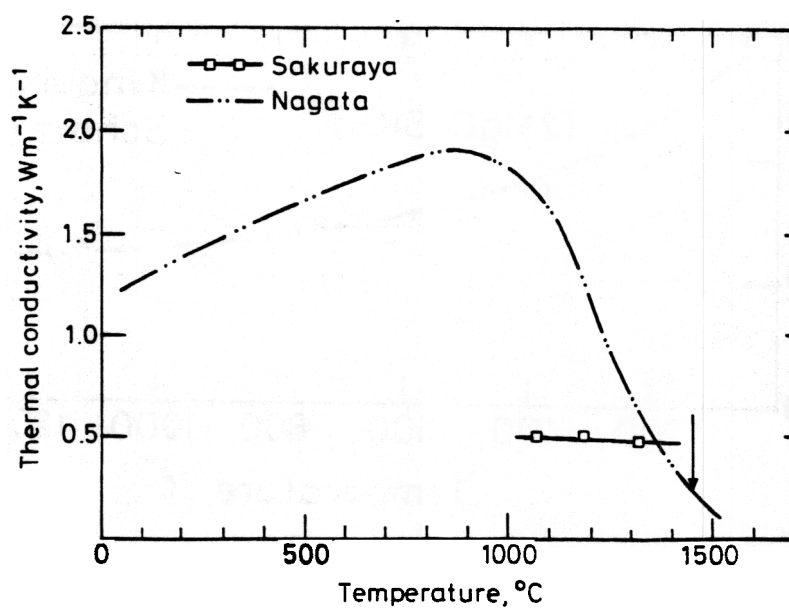
CaO-SiO₂

Figure 13 Thermal conductivity of slags with composition 55% CaO - 45% SiO₂ as a function of temperature (20,35).

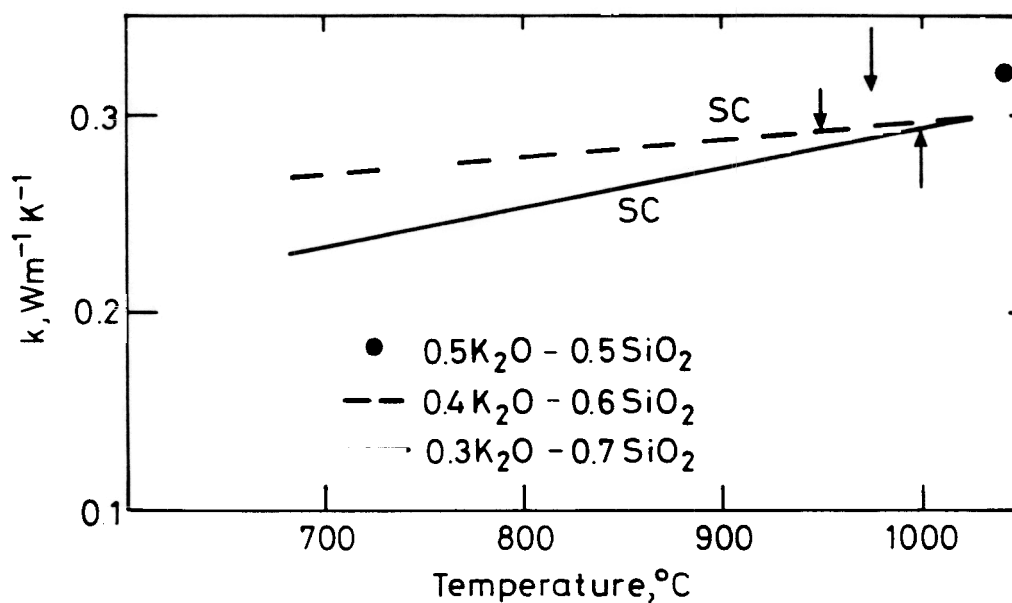
K₂O-SiO₂

Figure 14 Thermal conductivity of K₂O - SiO₂ melts as a function of temperature (18); arrows indicate liquidus temperatures and sc the supercooled liquid state.

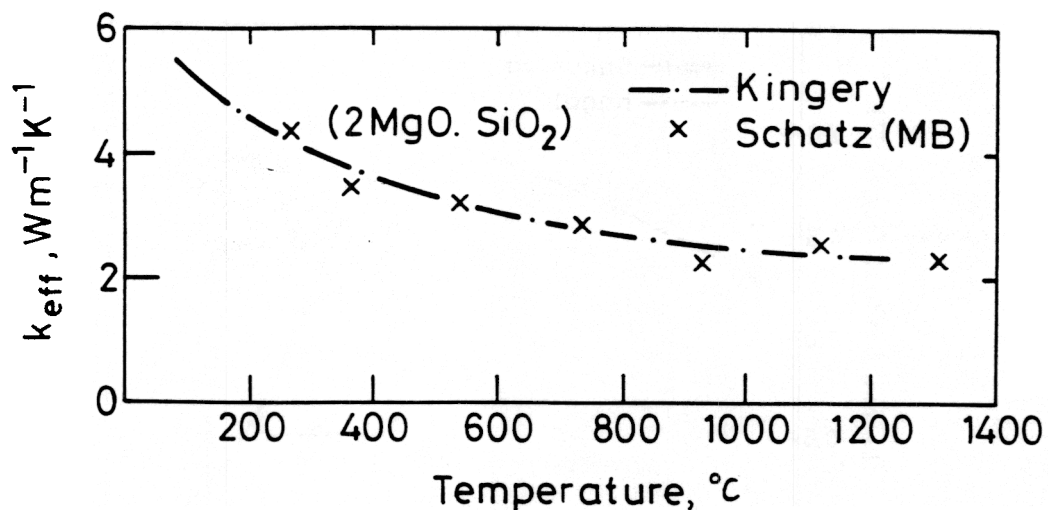


Figure 15 Thermal conductivity of fosterite (2MgO.SiO₂) as a function of temperature (10,15).

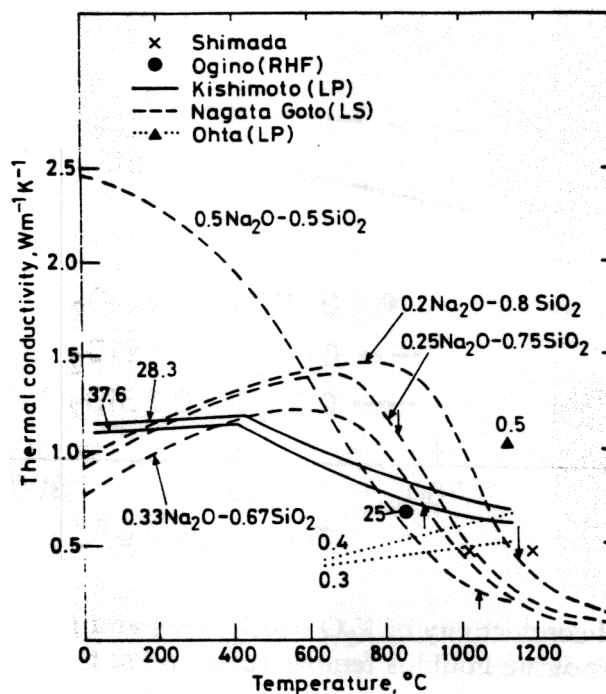


Figure 16 Thermal conductivity of Na₂O-SiO₂ slags as a function of temperature (11,18,19,20,36); compositions expressed in mole fractions and mass %).

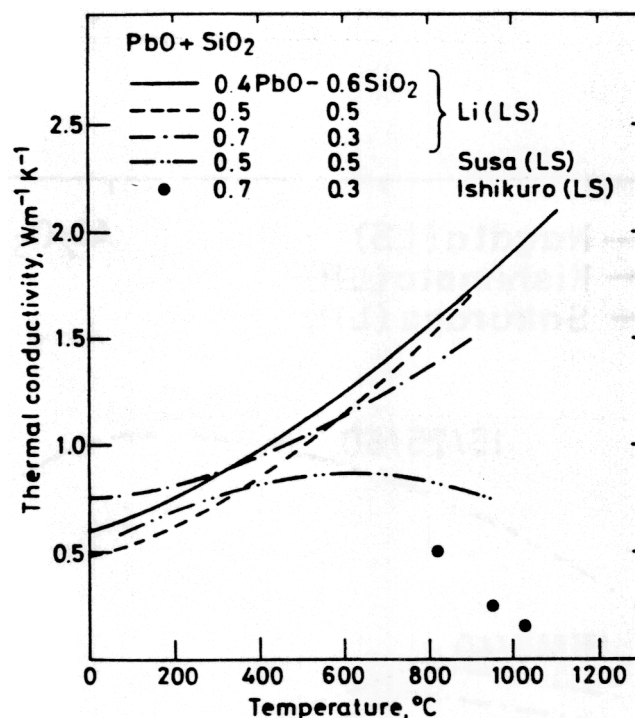
PbO-SiO₂

Figure 17 Thermal conductivity of PbO-SiO₂ slags as a function of temperature (37-39).

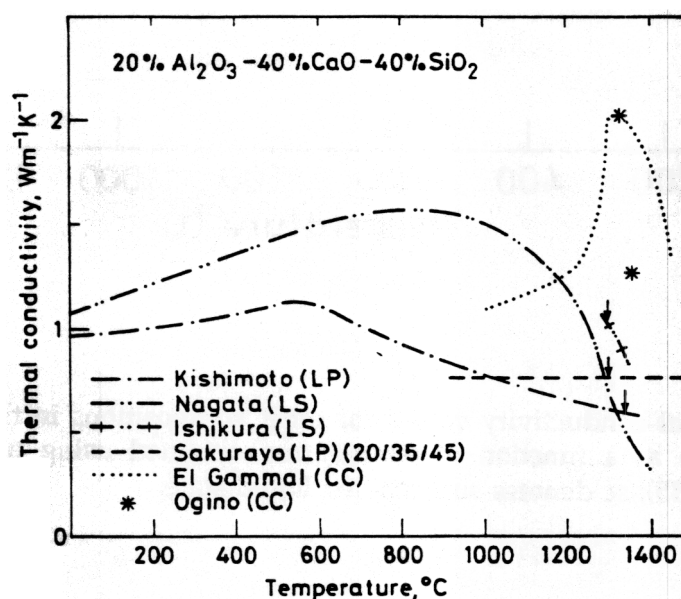
Al₂O₃-CaO-SiO₂

Figure 18 Thermal conductivity as function of temperature for slags 20% Al₂O₃ - 40% CaO - 40% SiO₂ (11,12,19,20,35,37); the values due to Nagata (20) and Kishimoto (19) are recommended, the differences in the values for the solid state may be due to differences in the relative amounts of crystalline and glassy phases. The k_{eff} values reported by Ogino (11) and El Gammal (12) probably contain significant radiation conduction contributions. Further references (32,40).

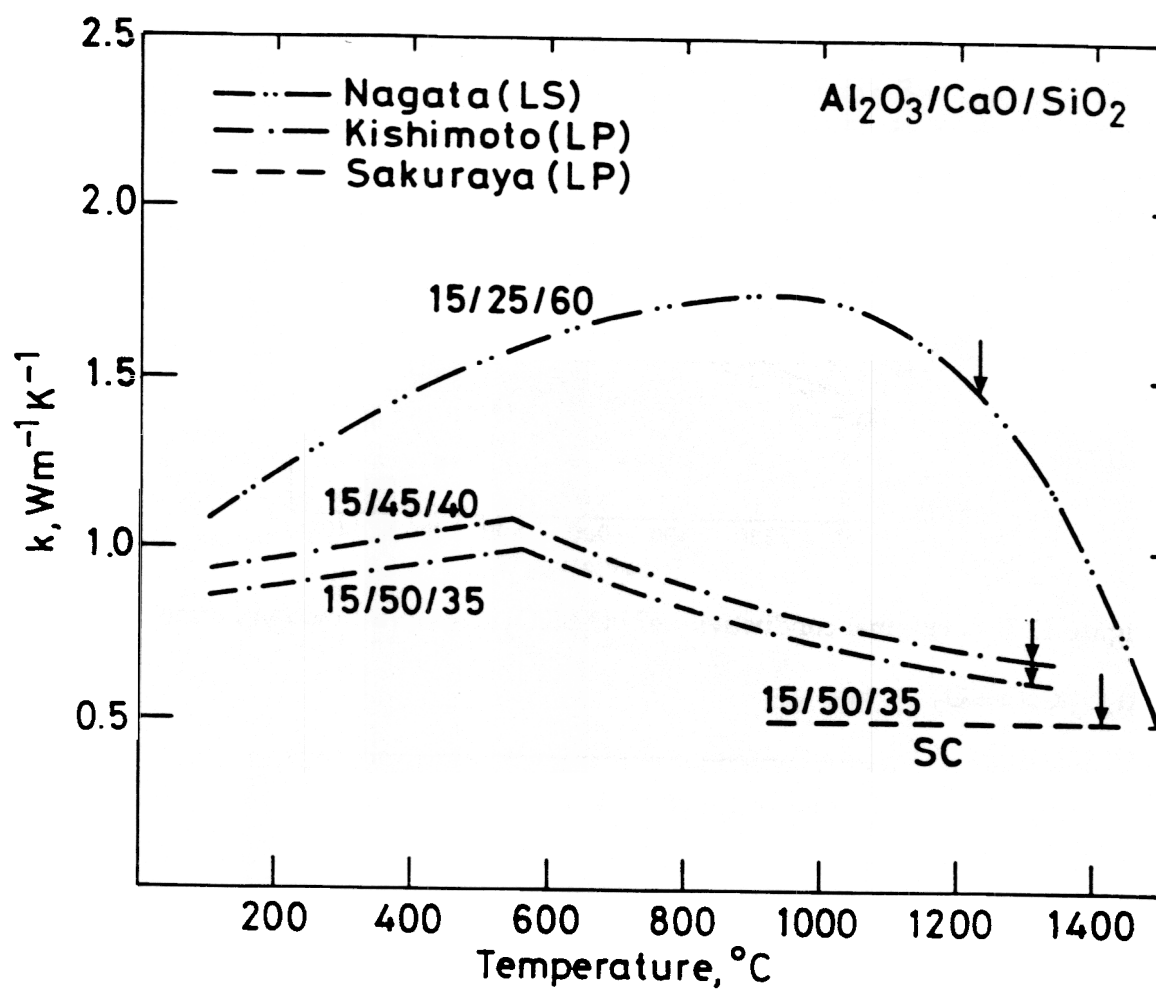


Figure 19 Thermal conductivity values for other compositions in the Al₂O₃-CaO-SiO₂ system as a function of temperature obtained using transient techniques (19,20,35); sc denotes supercooled liquid state.

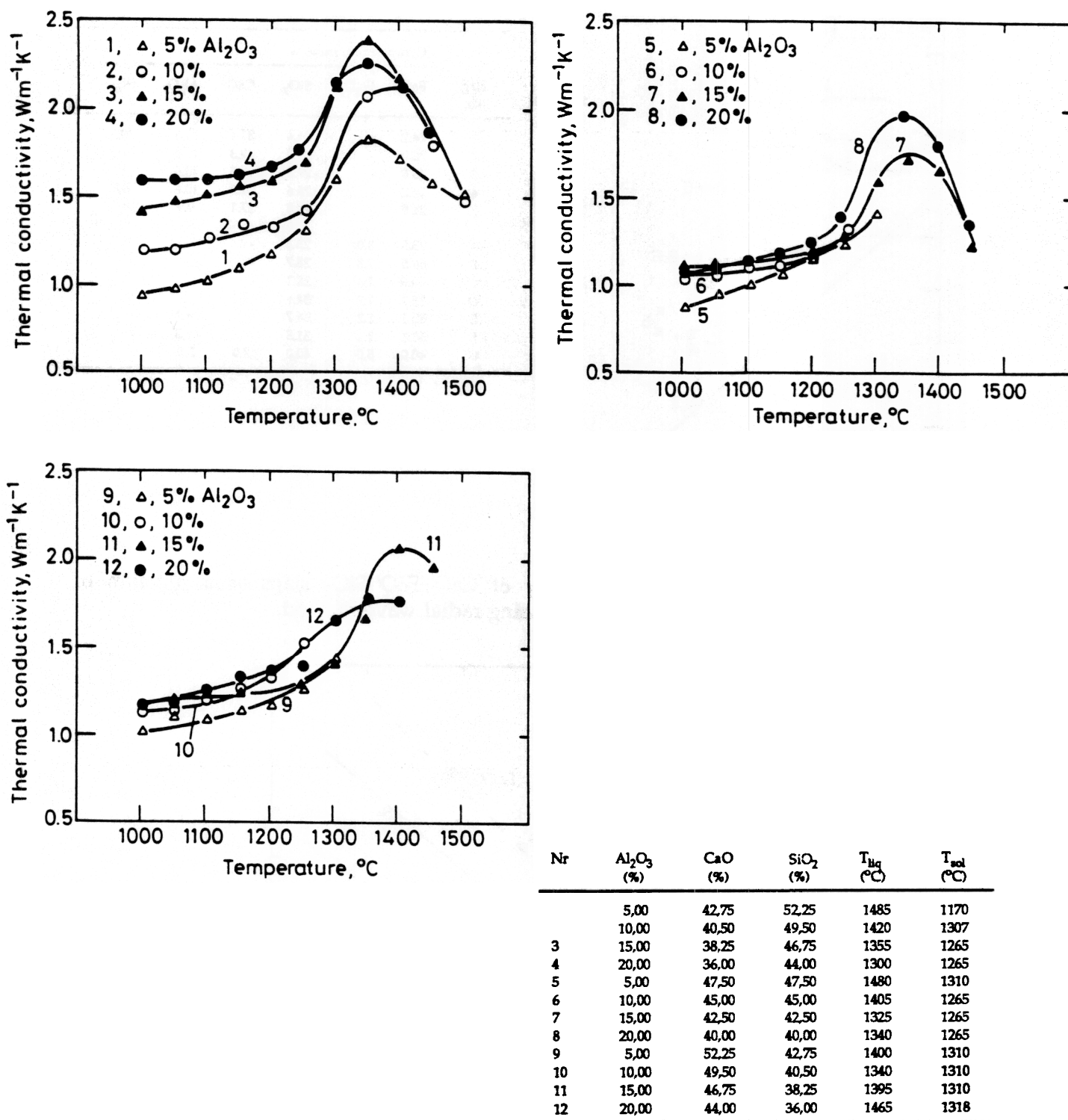


Figure 20 Thermal conductivity (k_{eff}) as a function of Al_2O_3 -CaO-SiO₂ slags obtained using (RHF) steady state techniques (12); these values probably contain significant contributions from radiation conduction.

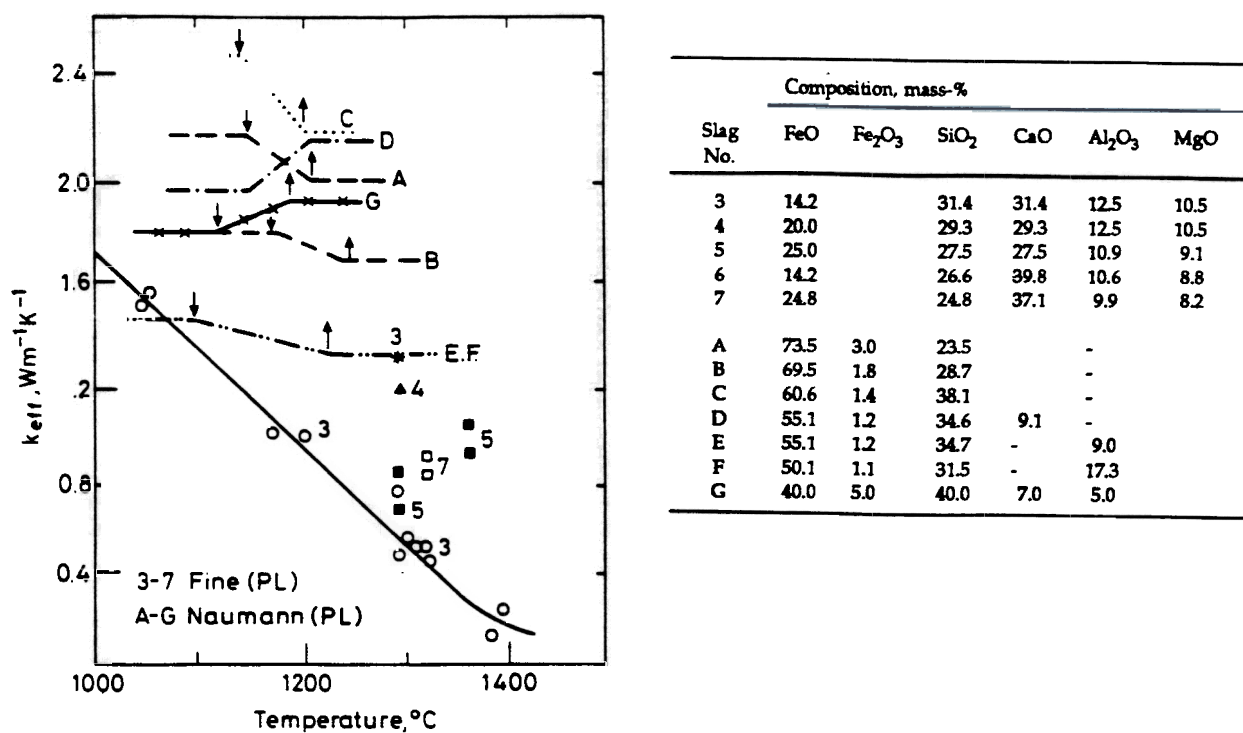
CaO-FeO_x-SiO₂

Figure 21 Effective thermal conductivity of CaO-FeO_x-SiO₂ slags as a function of temperature (13,14) obtained using radial wave method.

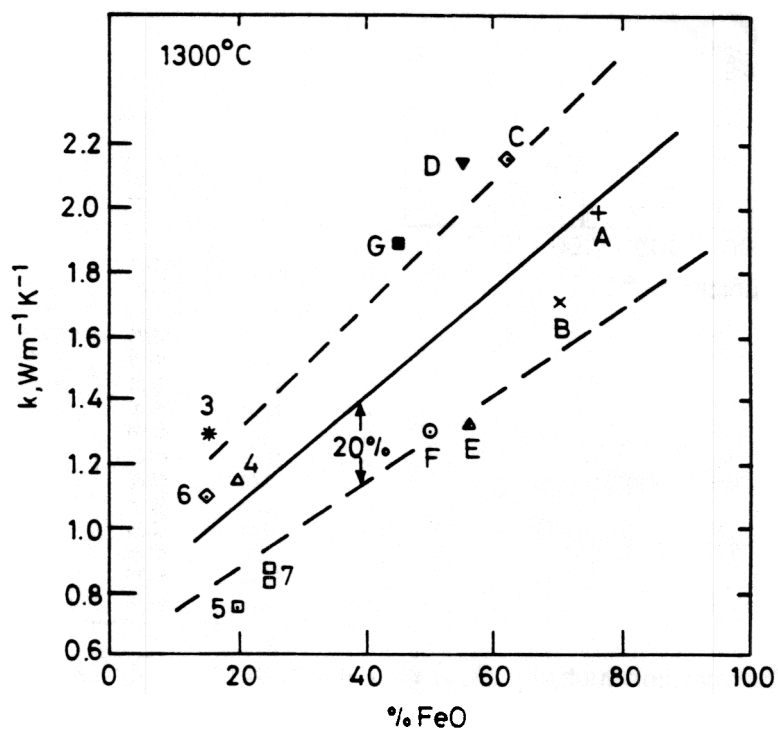


Figure 22 The effect of FeO content on the thermal conductivities (13,14) of CaO-FeO-SiO₂ slags at 1300 °C (41).

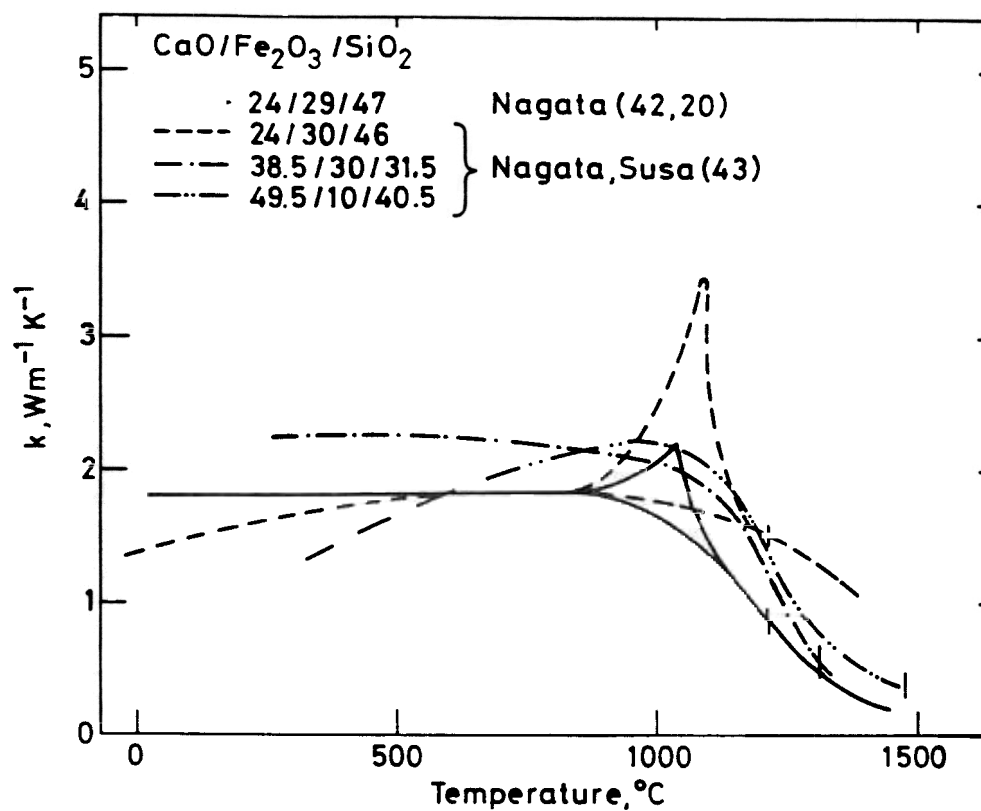


Figure 23 Thermal conductivity of CaO-Fe₂O₃-SiO₂ slags as a function of temperature (20,42,43); the (Fe²⁺/Fe³⁺) ratio was ca (0.2-0.3) in these slags (43).

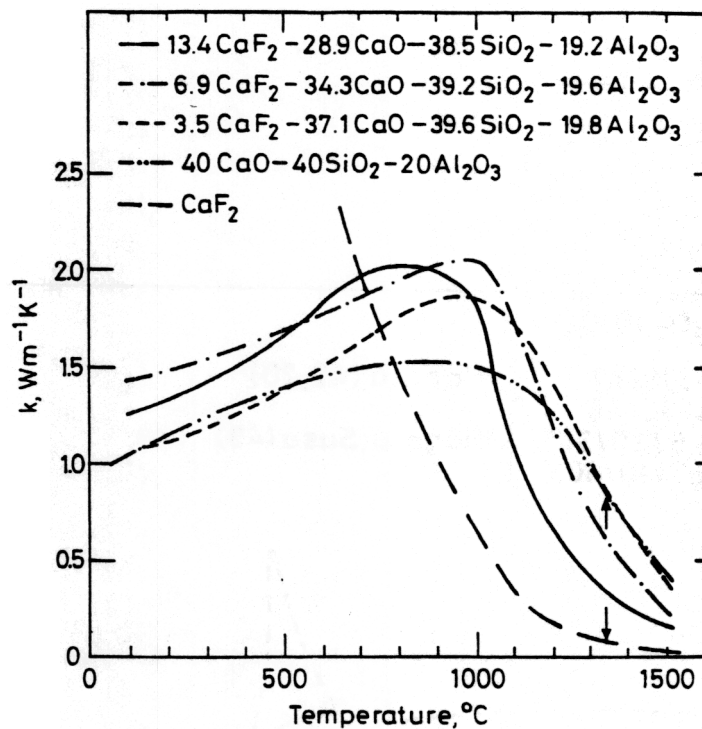
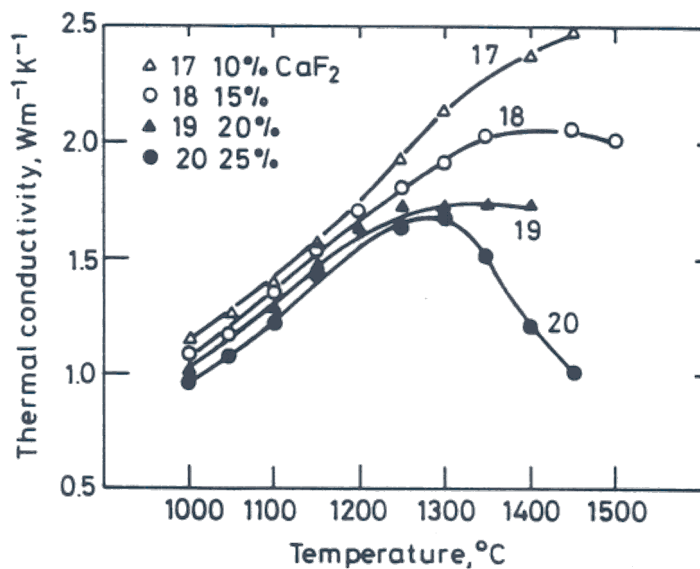
$\text{Al}_2\text{O}_3\text{-CaF}_2\text{-CaO-SiO}_2$ 

Figure 24 Thermal conductivity of $\text{Al}_2\text{O}_3\text{-CaF}_2\text{-CaO-SiO}_2$ slags as a function of temperature obtained with the line source method (20).



Slag No.	Mass %			
	Al_2O_3	CaF_2	CaO	SiO_2
17	13.5	10	34.4	42.1
18	12.8	15	32.5	39.7
19	12.0	20	30.6	37.4
20	11.3	25	28.7	35.1

Figure 25 Thermal conductivity of $\text{Al}_2\text{O}_3\text{-CaF}_2\text{-CaO-SiO}_2$ slags a function of temperature obtained using radial heat flow method (12); these results probably contain a contribution from radiation conduction.

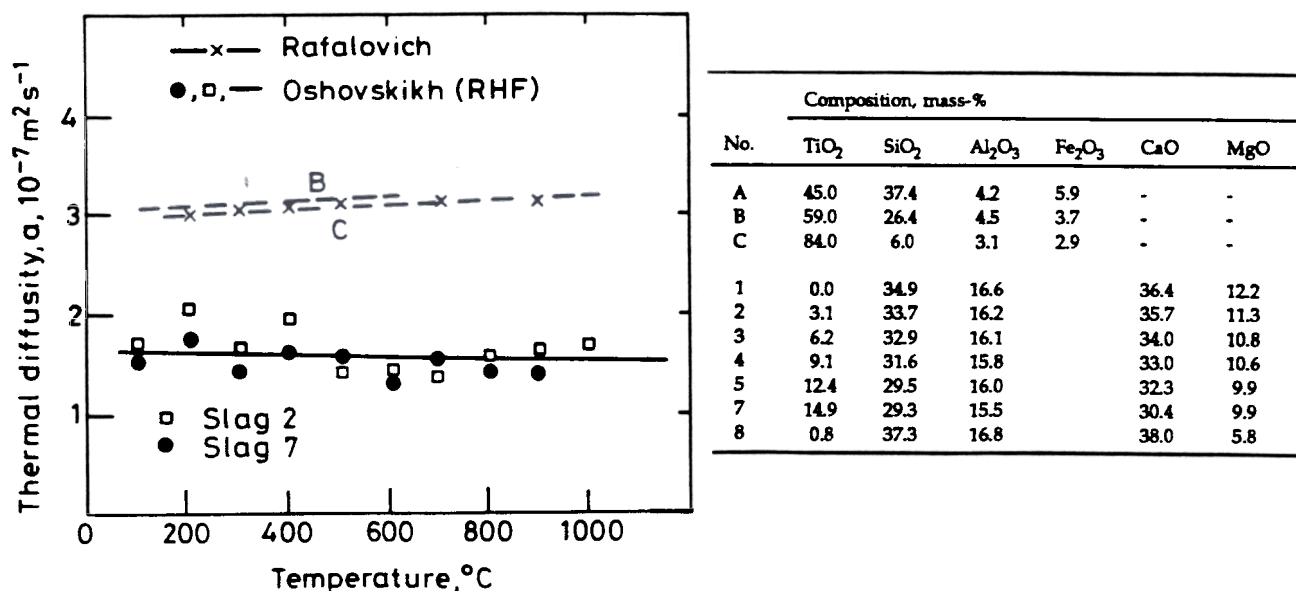
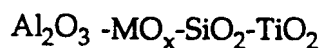


Figure 26 Thermal diffusivity of $\text{Al}_2\text{O}_3 - \text{MO}_x - \text{SiO}_2 - \text{TiO}_2$ slags as a function of temperature (32,44).

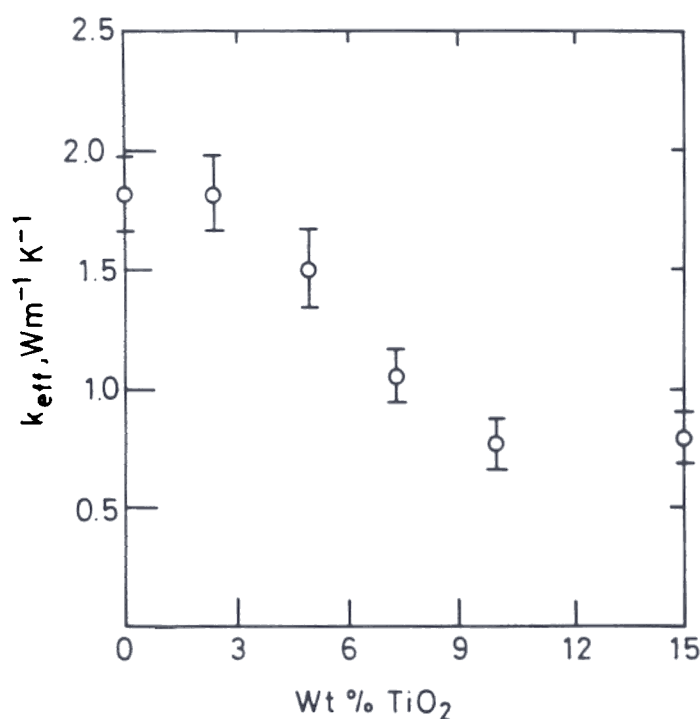
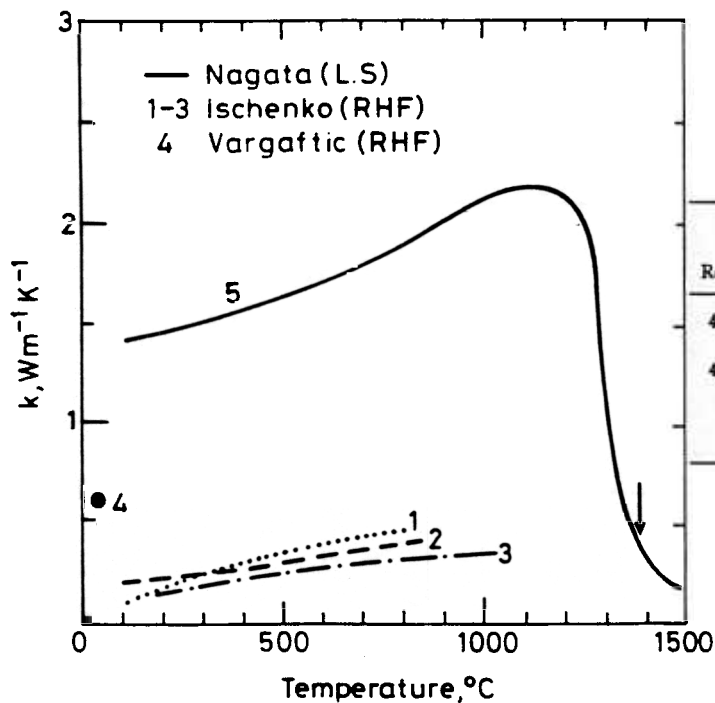


Figure 27 The effect of TiO_2 additions on the thermal conductivity of a slag containing 20% Al_2O_3 - 40% CaO - 40% SiO_2 (40); the decrease in k_{eff} was attributed to a decrease in k_R with increasing TiO_2 (40).

Blast furnace slags



Ref.	No.	Composition, mass-%						
		CaO	SiO ₂	Al ₂ O ₃	MgO	Na ₂ O†	FeO‡	Fe
45		Not given						
46	1	25.0	20.2	3.9	1.8	14.4	5.8	14.9
	2	18.1	11.1	2.2	1.9	25.1	4.2	45.7
	3	28.0	27.9	3.3	3.0	16.2	4.3	14.6
	5		41.7	34.5	6.1			

Figure 28 Thermal conductivities of blast furnace slags as a function of temperature (20,45,46); values shown as a solid line (20) are recommended.

Coal Slags

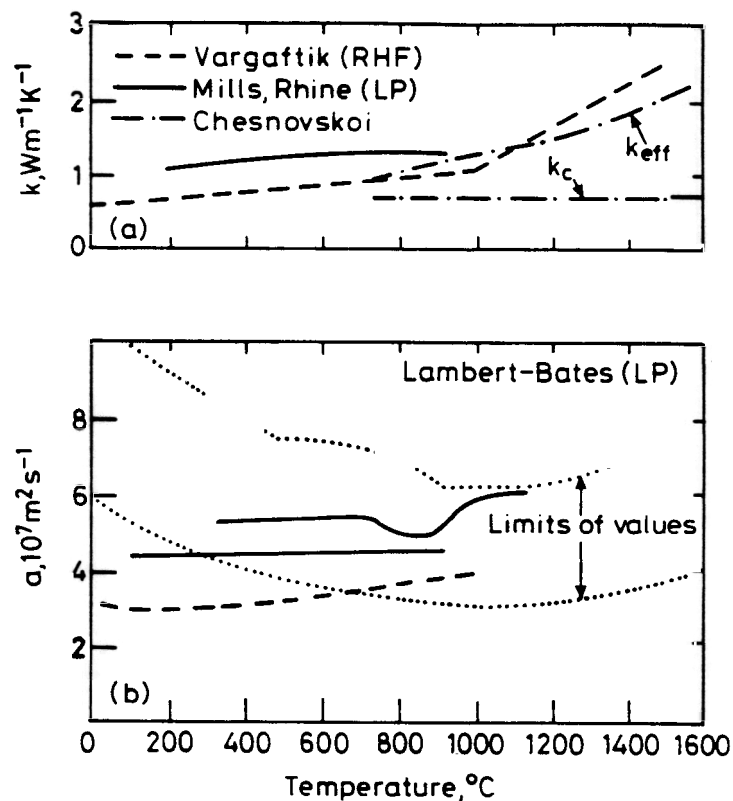


Figure 29 (a) Thermal conductivity and (b) Thermal diffusivity as function of temperature (45,47-50); Chesnovskoi (49) assumed a constant value for the lattice conductivity for all temperatures $> T_g$.

Continuous Casting Powders and Slags

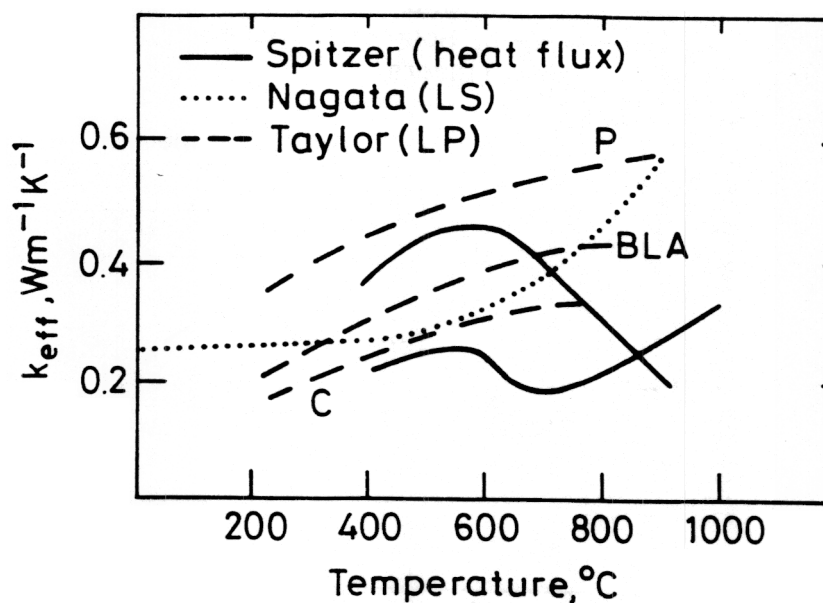


Figure 30 Thermal conductivity of powder form of continuous casting fluxes as a function of temperature (20,51,52); powder P (52) was in granulated form, all others were in the powder form.

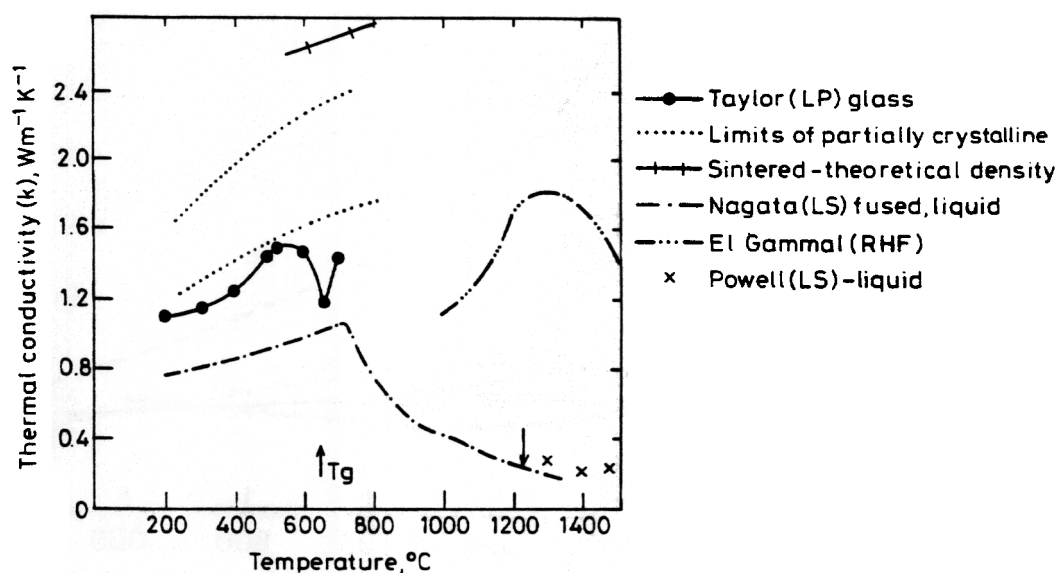


Figure 31 Thermal conductivities of continuous casting slags in the glassy and crystalline states as a function of temperature (12,20,52,53); the values reported by El Gammal (12) probably include contributions from radiation conduction.

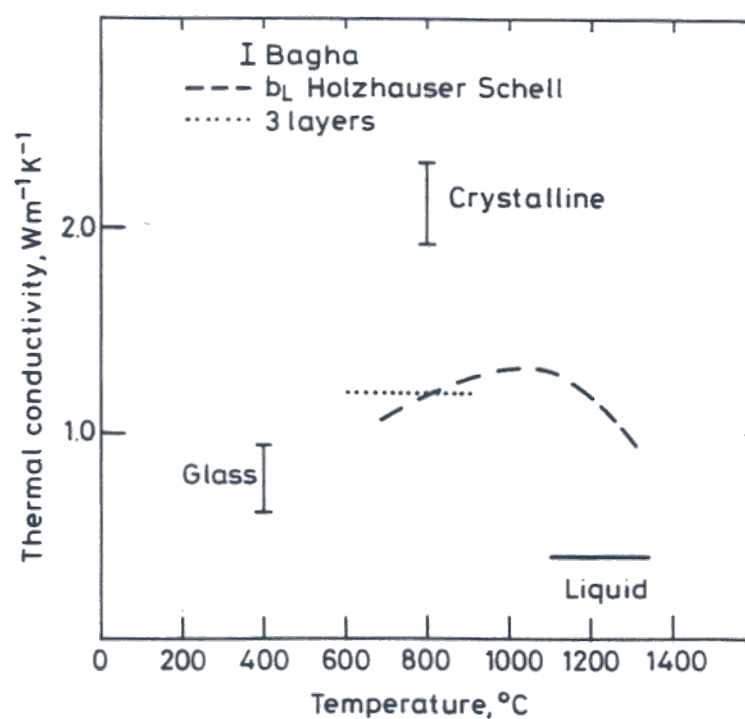


Figure 32 Thermal conductivity of continuous casting slags as a function of temperature as determined by analysis of heat flux measurements obtained in simulation tests (54-57); the results are in agreement with results shown in Figure 31. Further reference (58).

ESR slags

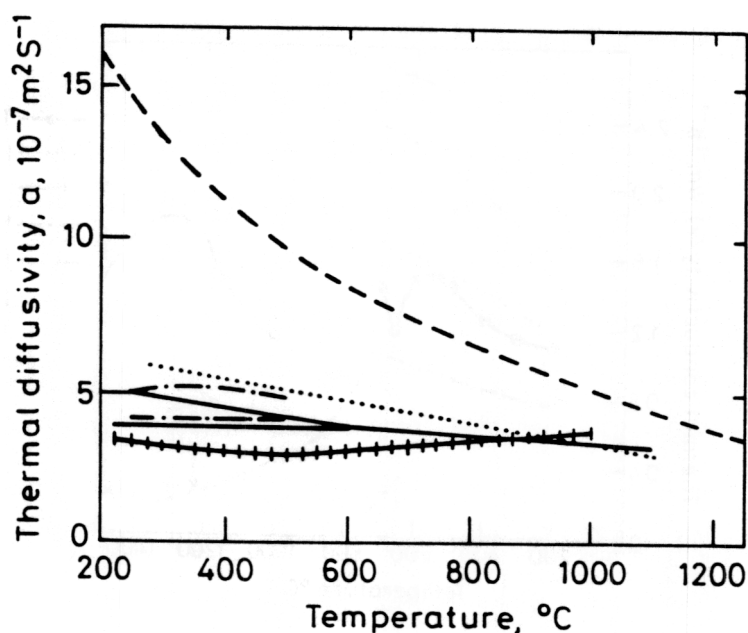
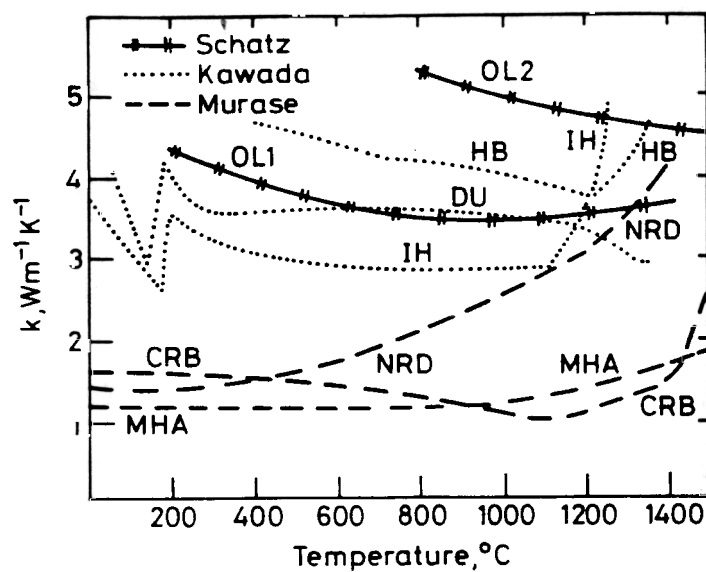


Figure 33 Thermal diffusivities of various (sintered) CaF_2 -based slags in Ar as a function of temperature (59,17); a value for the thermal conductivity k of $0.4 \text{ Wm}^{-1}\text{K}^{-1}$ has been reported for liquid CaF_2 - 15% Al_2O_3 - 15% CaO (28).

Rocks, Minerals



Ref.	No.	Composition, mass-%				
		SiO ₂	MgO	CaO	FeO	Al ₂ O ₃
59	HB			-	6.0	
				0.5	7.0	1.5
	IH			4.0	5.0	4.0
	DU			-	13.0	(5% Cr ₂ O ₃)
60 (RHF)	NRD	74.0	0.5	1.5	2.4	14.0
	LRB	60-75		Low		(+ 8% Na ₂ O + K ₂ O)
	MHA				15-25	
	g	Contract				
15 (MB)	01	40.8	49.3		9.9	
	02	41.6	52.7		5.7	
	DU	44.3	53.9		1.8	

Figure 34 Thermal conductivities of rocks and minerals as a function of temperature (15,59,60).

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