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THEME

Numerical Study of the Adaptation of Directional Solidification Furnaces for the Production of Mono-like Ingots

Publicly defended before the jury composed of:

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Dedication

I have the great pleasure of dedicating this modest work to my dear mother, who always gives me the

hope of living and has never ceased to pray for me.

To my dear father, for his encouragement, his support and his sacrifices so that nothing hinders the progress of my studies.

To my brothers, Walid, Youcef, Khalil, Amdjad, Mehdi. And all my dear teachers.

To my best friends Romissa, Maria, Oumlkhér and Hanaa.

And my dear colleagues and all those who have helped me to accomplish this modest work.

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I wish to thank all my professors, staff and technicians at the University of Laghouat for their co-operation, indirect or direct contribution in finishing my master degree.

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List of Abbreviations and Symbols

DS	Directional Solidification
CZ	Czochralski
KY	Kyropoulos
FZ	Float Zone
HEM	Heat Exchange Method
SoG-Si	Solar Grade Silicon
MG-Si	Metallurgical Grade Silicon
UMG-Si	Upgrade Metallurgical Silicon
EG-Si	Electronic Grade silicon
Mo-Si	Monocrystalline Silicon
Mc-Si	Multicrystalline Silicon
UV	Ultraviolet
IR	Infrared
PV	Photovoltaic
ppm	part-per-million
ρ	Density
C	Specific heat
p	Thermal sources heat
$\boldsymbol{\lambda}$	Thermal conductivity of
$\boldsymbol{K_S}$	Thermal conductivity of solid
$\boldsymbol{K_L}$	Thermal conductivity of liquid
$\boldsymbol{G_S}$	Thermal gradient in solid
$\boldsymbol{G_L}$	Thermal gradient in liquid
$\Delta \boldsymbol{H_F}$	The variation of the fusion enthalpy per mole
$\boldsymbol{V_S^M}$	Molar volume of the material
V	Normal speed
L	Latent heat
HPA	High Purity Alumina

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General Introduction

This numerical work is related to the study of the adaptation of industrial Directional Solidification (DS) furnaces for the production of mono-like crystalline ingots of Silicon and Sapphire materials.

Manufacturers are looking of monocrystalline ingots with high quality, but the problem is the cost of the production of monocrystalline ingots, it is very expensive because they use an expensive process method known as Czochralski (CZ) method.

The idea is to use a cheap method, like DS method that is used for the production of multi-crystalline ingots, and adapt this method for production of mono-like ingots with a close quality of monocrystalline products.

To materialize this idea, we are proposing new changes in the standard DS furnaces, based on the numerical simulation of thermal configuration of the DS furnaces before and after adaptations.

In chapters one and two, we present the context of this study, a general review on Silicon and Sapphire raw materials and finished products and the most process method that are used for their production.

The last chapter develops the numerical simulation of the DS furnace with Multiphysics software.

The simulation results are presented first, for the standard furnace configuration used in the industry, and second, for the standard configuration with the adaptations to address the issue of the possibility of producing mono-like ingots with DS method for both materials Silicon and Sapphire.

The numerical simulation results allowed us to offer interesting improvements that make it possible to adapt the standard DS furnace for mono-like ingots production.

Chapter 1

Introduction

1.1 Thesis Context

At nowadays, the world's economic activity growth lead to the augmentation of energy demand causing the resource shortage and environmental pollution due to the extensive consumption of conventional fossil energy resources (oil, gas, coal and wood).

All world governments are developing a clean and renewable energy sources (solar energy, biomass energy, hydropower energy, wind energy etc.) and innovative process for low energy consumption.

The Silicon is a semiconductor material used for solar-cell production, and the Sapphire is Alumina material used for numerous applications, as an example: LED and laser technology are very strategic materials.

Due to the high demand on these materials, extensive studies on Silicon and Sapphire production have been carried out over the world in order to obtain cheap materials with acceptable quality.

In the market, the final material products of Silicon and Sapphire are as a monocrystalline or multicrystalline crystal, the most important element that the two materials

have in comment that they can be produced by the same method process.Many methods are widely used to produce them, such as the Czochralski (CZ) method, the Kyropoulos (KY) method, the Heat Exchanger Method (HEM) and the Directional Solidification (DS) method.

DS and HEM methods are used for multicrystalline production; they play a leading role in the manufacture of various materials, they use the directional solidification physical principal for the process production.

In these two methods, the melting and crystallization processes take place as the same. The only difference lies in the way in which the crystallization occurs after the feedstock has been melted; in DS method, the crystallization is due to a mechanical movement of the crucible towards the cooler area, while in the HEM method the crystallization is due to a controlled heating power and a preferential heat extraction.

1.2 Thesis objectives

The best efficiency of Silicon and Sapphire final products are obtained from monocrystalline ingots, as an example we can see in Figure 1.1 the efficiency of Silicon solar cells comparison between the monocrystalline and multicrystalline products.

But it is highly expensive to produce a monocrystalline than multicrystalline ingot due to the use of CZ process; to have a clear picture;Table1.1 shows an interesting comparison of the most important parameters between the CZ and DS process:

The question this thesis answering is why we do not use a cheap process as DS process and produce in middle, between monocrystalline and multicrystalline crystal, ingot products to be used in applications when we do not have a high-quality expectation.

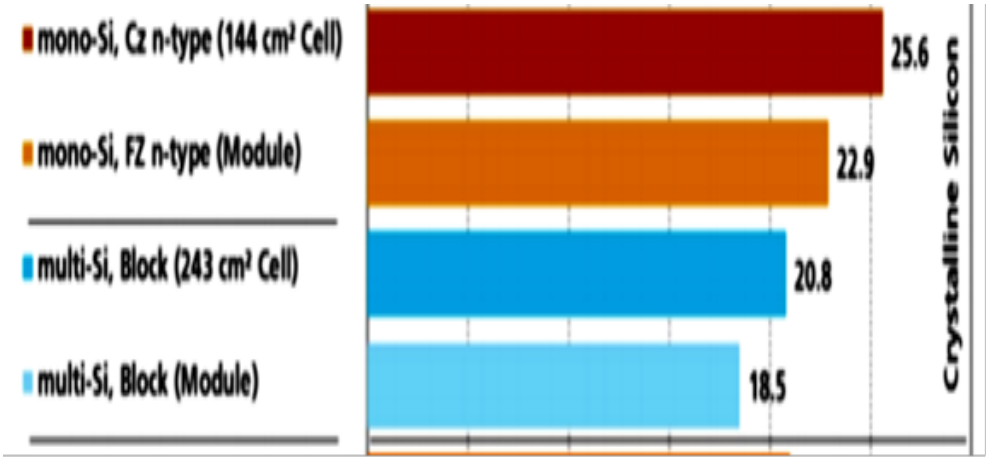


Figure 1.1: Efficiency comparison between Best Lab Cells and Best Lab Modules [22].

Tableau 1.1: Comparison between CZ and DS growth methods for solar Silicon.

Characteristics	**CZ**	**DS**	**References**
Crucible material	Quartz	Quartz	[12]
Consumable cost	High	Low	
Production by hour (PPH)	Low	Very high	
Production size (mm)	150 - 200	1000	
Operation skill	Less	Easy	
PolySilicon feed	Chunk	Chunk	
Seed size (mm^2)	$< 10 \times 20$	156×156	
Growth rate (cm/h)	4.5 - 9.0	$1.2 - 1.8$	[11]
Diameter or size (cm)	15-20	$69 \times 69, 84 \times 84$	
Crystal structure	Single	Multi or single	
Feedstock requirement	High/moderate	Low	
Solar cell efficiency	17-23	16-19	

The overall goal of this thesis is to study using a numerical simulation the possibility to produce an acceptable quality of Silicon or Sapphire ingots as mono-like ingots cheaply by optimizing the heat configuration due to the control process and adding new modifications to the standard DS furnaces.

Chapter 2

Silicon and Sapphire Materials & Processes

2.1 Silicon & Sapphire Materials

2.1.1 Silicon Raw Materials

Silicon is the most earth-abundant element; its melting temperature is around 1685 K. In nature, we find Silicon in the form of silicates, zircon, jade or quartz with different grades caused by the level of impurity. Due to the wide range of possible uses, Silicon has many industrial applications for solar cells field, despite the lower production costs for cells of certain materials (e.g. telluride cadmium, cadmium sulfide), the abundance, stability, non-toxicity, the efficiency and the control of its technology makes it a promising material for the future of the photovoltaic (PV) industry.

The first process in Silicon production is the carbothermic reduction of quartz.The end product of this step is known as Metallurgical Grade Silicon (MG-Si) and has the purity of 90%.

The second steps are the physical and chemical purification process. The end product of those processes is known as Upgraded Metallurgical Grade Silicon (UMG-Si) which

is MG-Si that has been further purified; but not quite to the purity levels of Electronic Grade Silicon (EG-Si). EG-Si is manly produced by the Siemens process, developed to produce material with less impurity for electronic industry [9].

To get an idea, Table 2.1 present the level of impurity for some grades of Silicon that exist in the market:

Tableau 2.1: Impurity concentrations in MG, UMG, SOG and EG Silicon (in ppm) [6].

Impurities	**MG-Si**	**UMG-Si**	**SOG-Si**	**EG-Si**
B	40	30	1	0.002
P	20	15	5	0.00à8
O	3000	2000	10	-
C	600	250	10	-
Fe	2000	150	10	0.01
Al	100-200	50	2	0.0008
Ca	500-600	500	2	0.003
Ti	200	5	1	0.003
Cr	50	15	1	0.003

2.1.1.1 Metallurgical Grade Silicon (MG-Si)

The mineral material used for the extraction of Silicon material is called "quartzite"; it is reduced in huge arc furnaces with carbon according to the following reaction:

$$SiO_2(s) + 2C(s) \rightarrow Si(l) + 2CO(g) \tag{2.1}$$

The Silicon obtained is called the Metallurgical Grade Silicon (MG-Si), is containing 1 to 2% of impurities.

2.1.1.2 Solar Grade Silicon (SOG-Si)

As presented in Table 2.2, depending on the producer and the routes used for the purification, we can find different grades of SOG-Si materials. SOG-Si or Upgraded Metallurgical

Grade Silicon (UMG-Si) is obtained from MG-Si using physicochemical purification processes as segregation phenomena of impurities, acid leaching and flux treatment.

Tableau 2.2: Impurity levels in different grades of SOG-Si materials [7] (in ppm).

Element	**MG-Si**	SOG - Si			
		Grade I	**Grade II**	**Grade III**	**Grade IV**
Al	$1200 - 4000$	< 0.001	< 0.02	< 0.3	< 1
B	$37 - 45$	< 0.001	< 0.02	< 0.3	< 1
P	$27 - 30$	< 0.001	< 0.02	< 0.05	< 0.72
Ca	590	< 0.01	< 0.05	< 0.1	< 4
K	-	< 0.01	< 0.05	< 0.1	< 4
Na	-	< 0.01	< 0.05	< 0.1	< 4
Cr	$50 - 140$	< 0.01	< 0.05	< 0.1	< 0.2
Cu	$24 - 90$	< 0.01	< 0.05	< 0.1	< 0.2
Fe	$1600 - 3000$	< 0.01	< 0.05	< 0.1	< 0.2
Mn	$70 - 80$	-	-	-	-
Mo	< 10	-	< 0.05	< 0.1	< 0.2
Zn	-	< 0.01	< 0.05	< 0.1	< 0.2
Ni	$40 - 80$	< 0.01	< 0.05	< 0.1	< 0.2
Ti	$150 - 200$	< 0.01	< 0.05	< 0.1	< 0.2
V	$100 - 200$	< 0.01	-	-	-
Zr	30		-	-	-
Sb	-		< 0.02	< 0.05	< 0.72
C	-		< 0.02	< 0.05	< 0.72
Mg	-	< 0.3	< 2	< 5	< 0.100

2.1.1.3 Electronic Grade Silicon (EG-Si)

The Electronics Grade Silicon is obtained after the purification of MG-Si using Siemens process. In this case, Silicon is oxidized to the trichlorosilane ($SiHCl_3$) compound according to the following reaction:

$$Si + 3Cl \rightarrow SiHCl_3 + H_2 \tag{2.2}$$

After, the Silicon is deposited as purified Silicon material. Although EG-Si is expensive, therefore the alternative metallurgical route is used to reduce the cost of obtaining a smaller quality of Silicon material. The choice of raw material is essential for the quality of the final product; in some case, the differences in final efficiency between EG and SOG-Si

is not higher than 1 % [6].

2.1.2 Silicon Final Product

One of the most important elements for the production of solar cells is the raw materials used for their production. As an example, for the monocrystalline ingots we use EG-Si and for the multicrystalline ingots we use the SOG-Si.

Figure 2.1: (a). Czochralski mono ingots, diameter: 800, length: 600 mm, (b). FZ mono ingot; (c). multicrystalline ingot [10].

There are two types of finished products, high quality monocrystalline Silicon ingots produced by CZ or FZ process (Figure 2.1 a, and b) and multicrystalline Silicon ingots produced by DS process with acceptable quality (Figure 2.1 c). Although there are a lot of variants of solar cells in the market, about 90% of them are made from Silicon, and the two most common types are monocrystalline cells (MO-Si) and multicrystalline cells (MC-Si).

2.1.2.1 Monocrystalline Silicon Cells

Made out from a cylindrical Silicon ingots (from CZ or FZ process) shaped design that helps optimize furnace performances. In this way, cells with monocrystalline have rounded

edges rather than square (Figure 2.2). The monocrystalline solar cells are the most efficient.

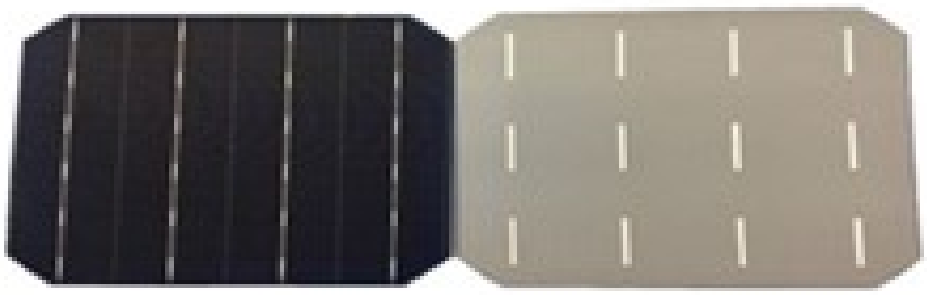


Figure 2.2: Monocrystalline Solar cells made for CZ or FZ ingots.

2.1.2.2 Polycrystalline Silicon Cells

Also known as multicrystalline Silicon cells were introduced to the industry in 1981. Instead, of using a cylindrical ingot they use a square ingot (Figure 2.1) that comes from DS process. However, polycrystalline cells are less efficient than monocrystalline cells.

Figure 2.3: Polycrystalline Silicon Solar Panel of SKTM, Laghouat, Algeria.

In Table 2.3, we summarize the main characteristics of each ingot material with the efficiency of the cell measured on solar modules and in the laboratory.

Tableau 2.3: Silicon ingots used for the production of photovoltaic cells [23].

Type of Silicon ingots	**Characteristics**	**Yield of Cells**	
		module	**Lab**
Monocrystalline (sc-Si)	Good yield, high energy production for cells	$18-22\%$	25%
Multicrystalline (mc-Si)	Good (performance / production) cost ratio, yield intermediate	$14-18\%$	20%

2.1.3 Solar Cells Prices

In Table 2.4, we show the prices announced by Energy Trend and PV Insight [24]. As we can see, mono cells are more expensive than multi-cells, due to the material and process costs.

Tableau 2.4: Solar cells Prices according to Energy Trend &PV insights in $/Watt [24]

Type of Solar Cell	PV Insights	Energy Trend
Multi-Si Cell	-	0.076
Mono-Si Cell	-	0.105
High Eff Mono-Si Cell	-	0.112
Superior High Eff Mono-Si Cell	-	0.123
Multi Cell Price	0.069	
Poly PERC Cell	0.093	-
Mono PERC Cell	0.104	-
China Mono PERC Cell	0.103	-

2.1.4 Sapphire Raw Materials

The raw material used for growing Sapphire ingot crystal is the Alumina (Al2O3). Sapphire is an oxide crystal with melting point around 2323K.

Figure 2.4 present the types of raw materials that we can find in the market.

It can be under a form of a scrunched powder, sintered charges, cracked crystal or polycrystalline ingots.

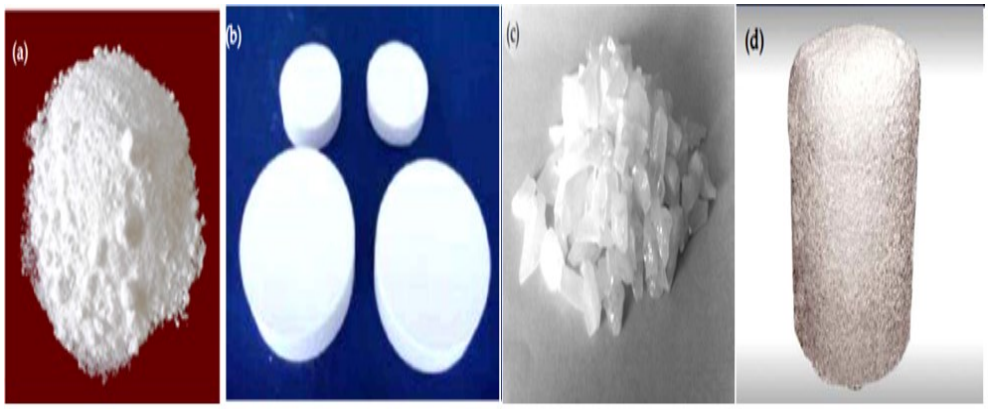


Figure 2.4: Sapphire raw materials: (a). Powder, (b). Sintered charge, (c). Cracked crystal, (d). Polycrystalline ingots [13].

In order to obtain a LED ingot grade Sapphire, the raw materials should be of a high purity level, at least $\geq 99.996\%$. Table 2.5 shows an example of high purity Alumina.

Tableau 2.5: Impurity levels in high purity Alumina [13].

Elements	Fe	Na	K	Ca
Content (mg/Kg)	< 4	< 1	< 1	< 1

2.1.5 Sapphire Final Product

Sapphire can be used at the highest temperature; the final products of Sapphire ingots are produced with methods that give a single (mono) crystal type as shown in the following Figure:

It has a high refractive index and a wide transmission band of 0.14 to $6.0\mu m$, covering the UV, visible and IR bands. The Sapphire also has a high hardness (next to the diamond) and a surface softness, very good tensile strength, low thermal conductivity, electrical insulation, resistance to wear, and resistance to thermal shock [12]. The combination of excellent optical and mechanical properties with high chemical durability makes Sapphire desirable material for LED and Laser applications

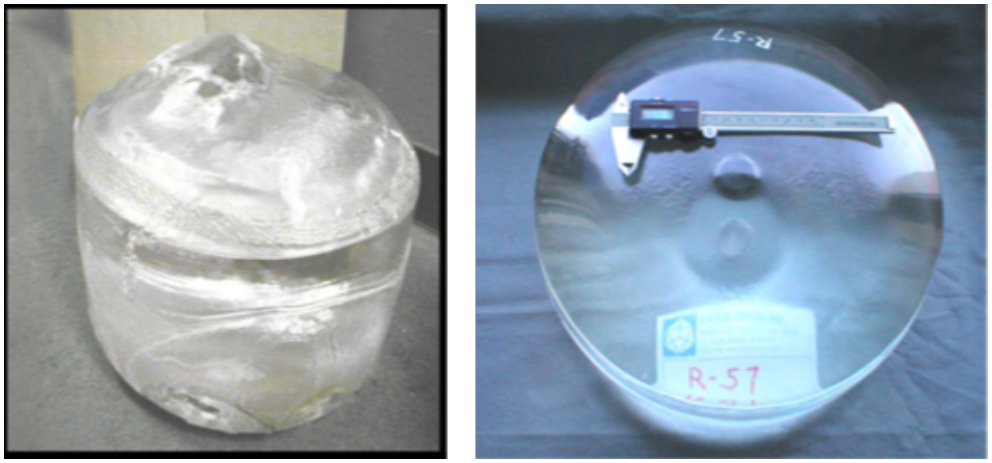


Figure 2.5: Mono crystal ingots of pure Sapphire [15].

2.1.6 High Purity Alumina Prices

Table 2.6 shows the prices of High Purity Alumina in the world. We can see the price increase with the quality of the material. The choice of the grades depends on the final quality we are looking for.

Tableau 2.6: High Purity Alumina Prices [25].

Grades	Europe	North America	Asia Pacific	Other	Global
4N	\$31-\$36	\$32-\$40	\$25-\$32	\$30-\$35	\$25-\$35
5N	\$50-\$60	\$55-\$60	\$48-\$55	\$50-\$60	\$48-\$60
6N	\$160-\$175	\$170-\$175	\$150-\$160	\$160-\$170	\$160-\$170

2.2 Furnaces and Production Processes

2.2.1 Czochralski Process (CZ)

The Czochralski process is the most used growth method for mono ingots. Taking its name from its inventor, it has been used for the development of materials such as Sapphire, Silicon, Germanium and Gallium Arsenide. It is preferred to other methods because the solid crystal is not in contact with the furnace elements, only a crucible used to contain the liquid material.

The quality of the single crystal obtained by this technique is linked to the thermal regulation; the principle consists in solidifying a single crystal by initiating growth from a bath of melted material by a monocrystalline germ of known orientation (Figure 2.6).

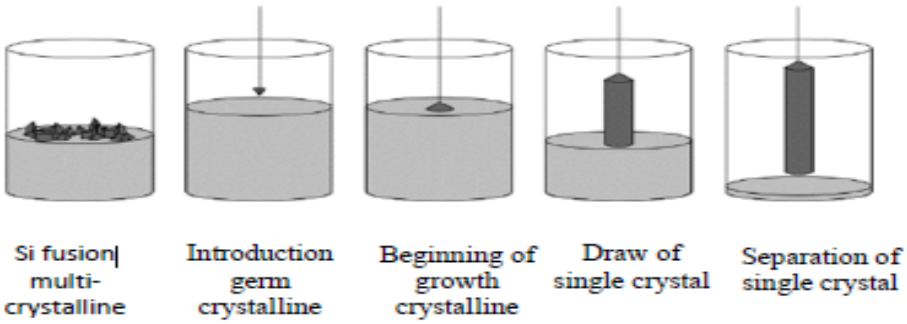


Figure 2.6: Czochralski process for Silicon production.

In the case, the melted material is made from high purity Silicon; the liquid is contained in a silica crucible (SiO2). A germ is put in contact with the bath and the draw is launched gradually and simultaneously with rotation.

The control of the rotation speeds defines the dimensions of the ingot. Currently, the largest ingot has a diameter of 450 mm for a height of 2150 mm and a yield of 940 kg.

The same technique is used to grow monocrystalline oxides like Sapphire. When a monocrystalline germ rotated on the surface of the molten material, by capillary action the liquid rises a few millimetres and remains attached. The germ, always in rotation, is then slowly pulled up. The interface is kept close to the surface of the bath, so that the extracted liquid solidifies and crystallizes on the germ (Figure 2.7).

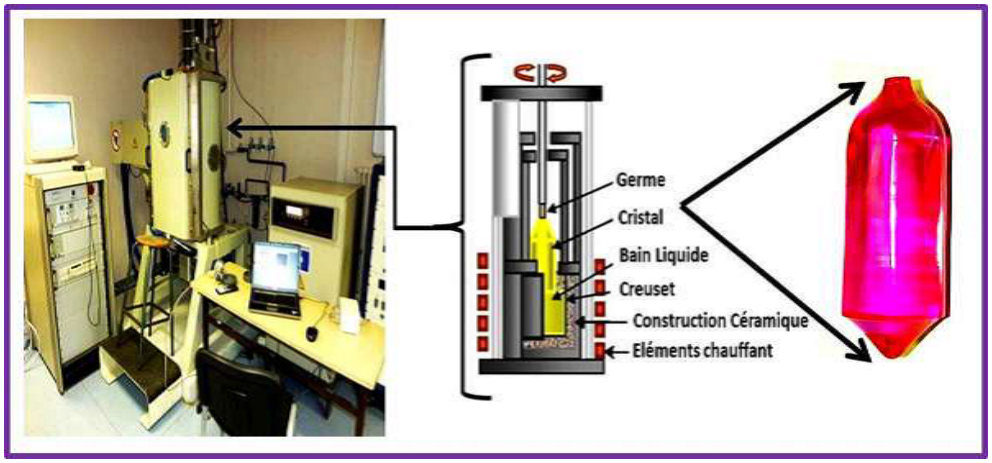


Figure 2.7: Czochralski method for sapphires production.

2.2.2 Float zone Process (FZ)

FZ process (Figure 2.8) gives monocrystalline ingots with the lowest density of impurities. For Silicon material, they use a polycrystalline Silicon raw material in form of a rod. In the processing, the end of the rod is heated up and melted using electromagnetic coil, the melted part is put in contact with a mono crystal seed, as the molten zone is moved along the poly-silicon rod, the single crystalline ingot is growing as well.

The advantage of FZ process is that the molten material is not in contact with other materials like quartz in CZ method.

The diameters of FZ ingots are generally not greater than 150 mm due to the surface tension limitations during the growth process. The FZ Silicon ingot is the best in quality [10].

The pictures below shows a FZ machine, only the upper part is visible, the total height of the machine is about 12 m. from Courtesy of PVA TePla [10].

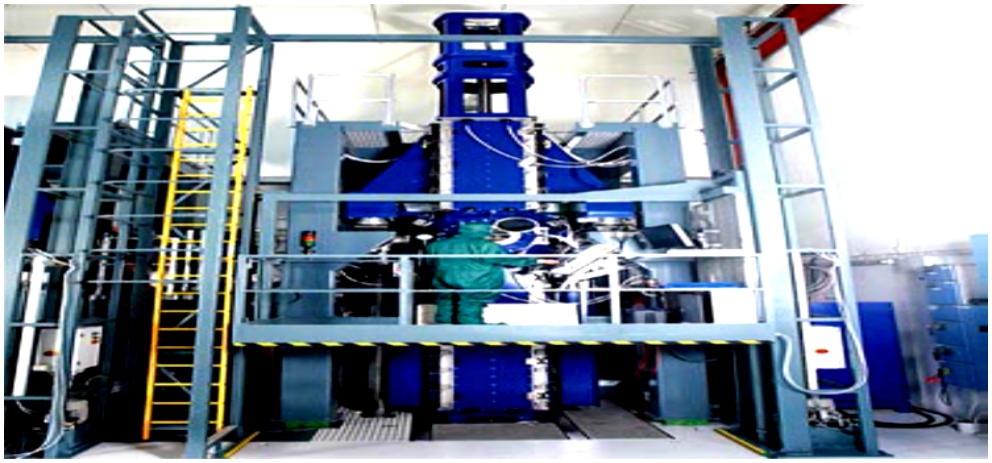


Figure 2.8: Float-Zone machine for silicon growth [10].

2.2.3 Kyropoulos Process (KY)

As CZ method, after bringing the monocrystalline germ into contact with the liquid the growth takes place by decreasing the power induced in the liquid bath as shown in Figure 2.9. In a well-designed system, the resulting crystal has a shape similar to the crucible but with a diameter less than or equal to 90% of the diameter of the crucible.

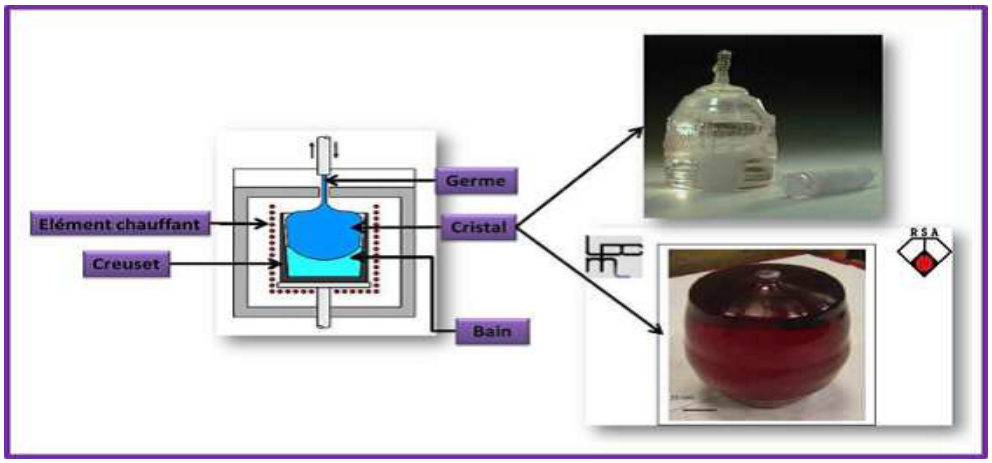


Figure 2.9: Principle of the Kyropoulos method: Sapphire and Sapphire doped with Titanium.

2.2.4 Heat Exchanger Method Process (HEM)

The HEM principle is to apply a heat exchanger to dissipate heat, making the crystal growth region forming a longitudinal cold / hot temperature gradient.

By controlling the volume of the gas flow inside the heat exchanger and changing the heating power, the gradient can be kept constant.

Then the crystal can grow slowly from the bottom to the upper part as shown in Figure 2.10 [15]. The HEM method is used for the sapphire to grow a large ingots size.

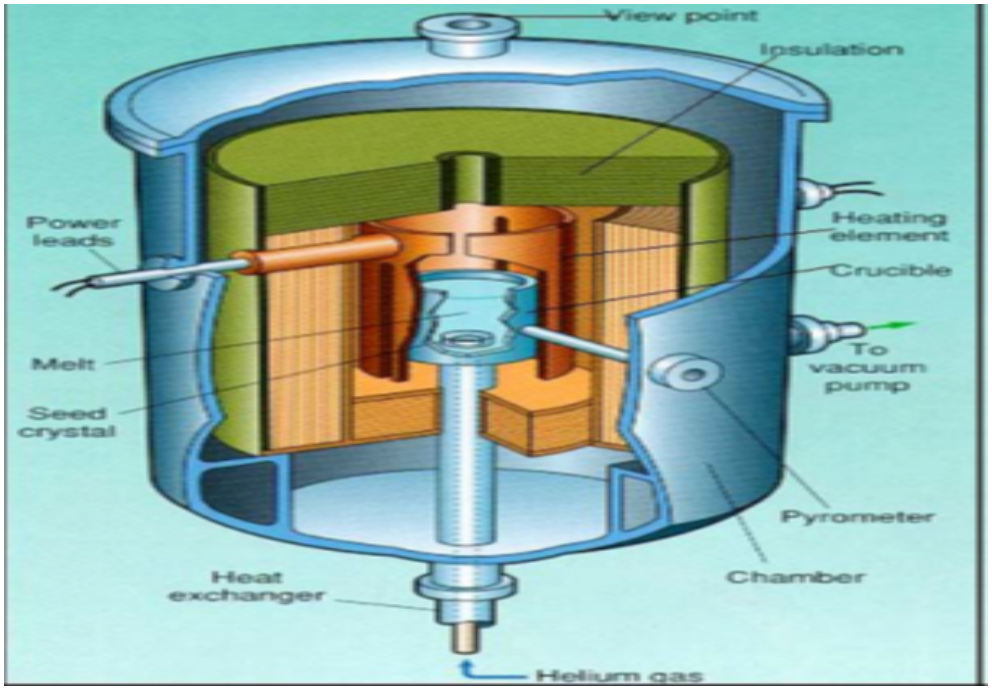


Figure 2.10: Principle of the Kyropoulos method: Sapphire and Sapphire doped with Titanium.

2.2.5 Directional Solidification Process (DS)

The DS process or the Bridgman process is a directed solidification technique; it consists of melting a charge of materials contained in a crucible and solidifying it in a controlled environment.

To obtain a directed solidification, the furnace should contain: adiabatic, cold and hot zones (Figure 2.11).

When a thermal gradient is applied, the solidification is performed either by moving the crucible with material from the hot zone to the cold zone, or by moving the adiabatic zone, or by controlled the cooling system.

The DS can be used to control the crystalline form and the purification process of the materials, this control is due to the control of the thermal gradient and the solidification speed. For Silicon, the DS is one of the most used processes for the growth of multicrystalline ingots for PV applications; it does not require a high quality of feedstock.

A square shaped ingot is used to avoid a large amount of material waste; during solidification, the heat is controlled in order to keep the liquid/solid interface between Silicon solid and liquid as flat as possible.

With DS method, we can use a crucible containing a feedstock material with the addition of monocrystalline seeds as germs in the bottom, the charge is melted with the top part of the seed, and then the crucible is moved in a gradient temperature for solidification at controlled speed and controlled growth.

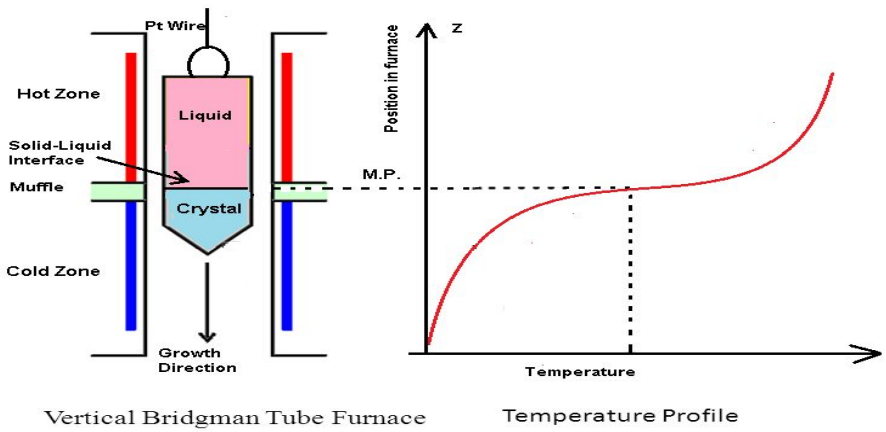


Figure 2.11: Principle of the vertical Bridgman method.

Chapter 3

Numerical Study of the Adaptation of DS Furnace to the Production of Monolike Ingots

3.1 Introduction

It is highly expensive to produce a monocrystalline ingot by the CZ process compared to the DS process. If we can use a cheap process like the DS process with high production to produce a quality ingot between CZ and DS qualities, it will be a huge improvement; because many applications do not need a high ingot quality like CZ quality, but they need better quality than DS ingots.

As we know, the efficiency of DS cells is lower than the CZ cells; but with monolike ingots, it is possible to get closer to the mono cells efficiency; this can be done by controlling the DS thermal field to improve the crystal quality growth, as in DS process the thermal field influences the interface shape, the temperature gradient, and the impurity distributions in the crystal.

The overall goal is to investigate numerically the possibility to producing acceptable quality of Silicon or Sapphire ingots as monolike ingots at lower costs by optimizing the

thermal control process and adding new modifications current DS furnaces.

3.2 Industrial DS Furnaces

The most widely used industrial DS furnaces in the world come from these different manufacturers (Figure 3.1):

- The French manufacturer: ECM,
- The American manufacturer: GT Solar,
- The Chinese manufacturers: JYT and JJL,
- etc.

All these furnaces use the same physical principle of directional heat extraction from the bottom with the hot and cold zone design; this design is suitable for the impurities segregation process and the crystal growth control. Their advantage, that they can process unique solidification growth with high capacities.

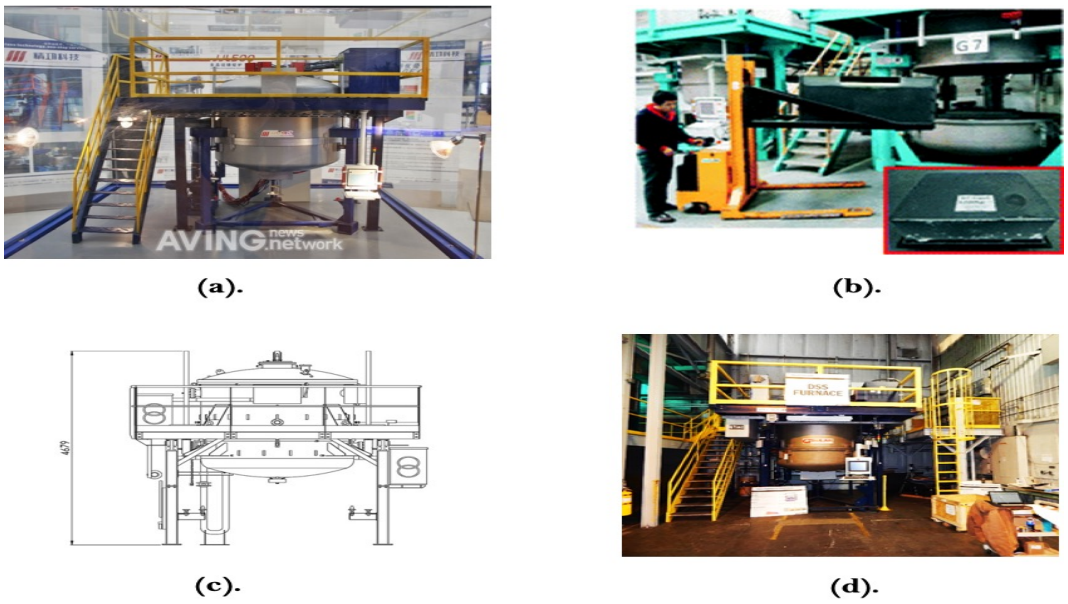


Figure 3.1: Pictures of industrial DS furnaces (a). JJL, (b). JYT, (c). ECM, (d). GT Solar [25].

3.3 Control Process of DS Furnaces

In practice, to have a high DS ingot quality, it is necessary to control all the process parameters, the most important controls used in industrial production are:

3.3.1 The Pulling Velocity Control

The quality of the ingot is related to the level of impurity and the crystalline form. These elements are related to the impurity segregation process and the crystal growth control related to the puling speed. The pulling velocity is imposed by the operator using the pulling motor of the DS furnace. To obtain high quality ingots, the pulling speed should be less than or equal to the theoretical solidification velocity (V) of the solidification process. This theoretical velocity is related to the thermal configuration generated to the design of the DS furnace and the operation parameters; it can be calculated by the following equation:

$$K_S G_S\text{-}K_L G_L = (\Delta H_F / V_M^S) V \tag{3.1}$$

This equation represents the exchanges of thermal energies at the solid/liquid interface. The interface moves at a normal speed (V), with a release of a heat due to the phase change.K_S and K_L are the thermal conductivity of solid and liquid phases of the material, G_S and G_L are the thermal gradients in the solid and liquid phases, ΔH_F represents the variation of the fusion enthalpy per mole, and V_M^S is the molar volume of the material.

3.3.2 The Thermal Control

In this case, the velocity of the solidification is related to the physics generated by the thermal filed inside the furnace. In this situation of thermal control, the thermal gradients in the solid and the liquid phases (equation 3.1) must be stabilized according to the solidification velocity wanted.

The two zones (hot and cold) of the furnace should be well controlled and well insulated

to avoid the heat loss. This stabilized thermal control is generated by the power control profile that is imposed by the operator (Figure 3.2).

As can be seen in Figure 3.2, we can impose the temperature profile by thermocouple is positioned TC1 (as an example, at the position of the top heater) and let the furnace control the power necessary to reach this temperature profile. Or, by imposing the power profile and recording the temperature at the TC1 thermocouple.

In both cases, the temperature and power profile are obtained by numerical study and validated by the experimentation.

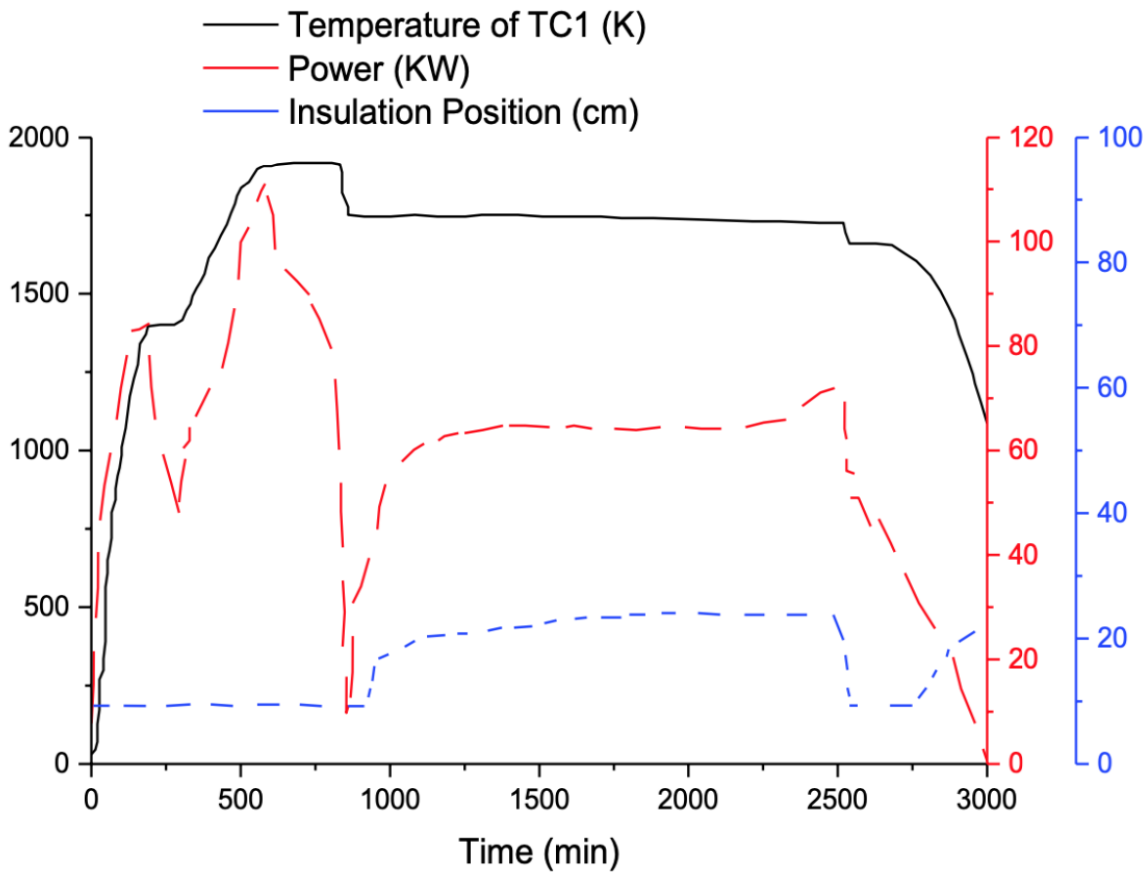


Figure 3.2: Power and Thermal control of industrial DS furnace [16].

3.4 COMSOL Multiphysics Software

COMSOL software is a multi-physical solver, which is in the form of a platform allowing modelling and simulating several physical phenomena. Its strong point is the coupling between different processes; more than 30 different physical modules allow researchers to simulate their process.

These modules offer specialized interfaces in acoustics, chemical engineering, geophysics, electromagnetism, heat transfer, transport of matter and momentum, and structural mechanics, etc.

In addition, COMSOL is equipped with an interface that can link the simulations carried out with other software.

3.5 Standard and Adapted DS Geometry

As an example for Silicon production, there are several sizes of Silicon ingots, from G1 to G8. The number 1 in G1 represents the number of the bricks in the side of the ingot after the cutting process, as shown in Figure 3.3; we present a G5 ingot with 5x5 bricks of 16x16 cm2.

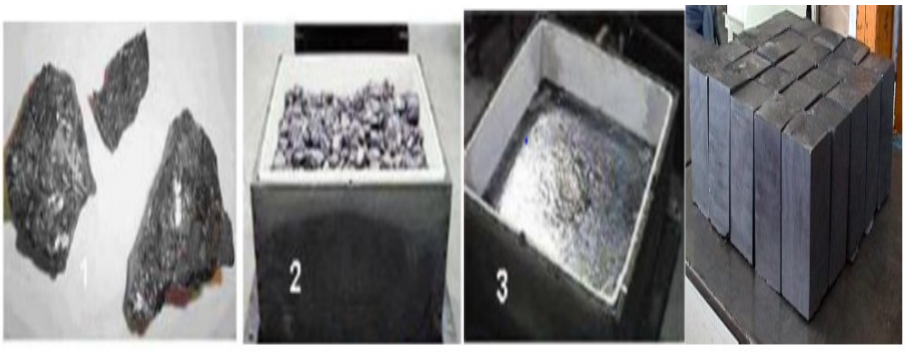


Figure 3.3: Standard processing of DS feedstock and cutting (G5 ingot: 5x5 bricks).

For our adaptation, before filing the crucible with Silicon chunks feedstock, it is necessary to put some seeds of monocrystalline silicon (2 to 3 cm thick) because these will be the germs that initiate the crystallisation of a monolike crystal. As shown in Figure 3.4:

In order to simplify the numerical simulation, the geometry of the DS furnace that is used in our study is presented in Figure 3.5:

In Figure 3.5, the furnace is made of double-wall steel shell enclosure, with water movement for the cooling system. Inside this enclosure, there is another graphite fibre enclosure to insulate the hot area and keep a stable temperature inside the furnace.

Heating elements made by solid graphite, in standard geometry, they are installed on the top and sides of the crucible holder (in solid graphite) and in adapted geometry; we will add a heating element on the bottom of the crucible holder. And finally, we have the crucible (in quartz) filled with a silicon material charge.

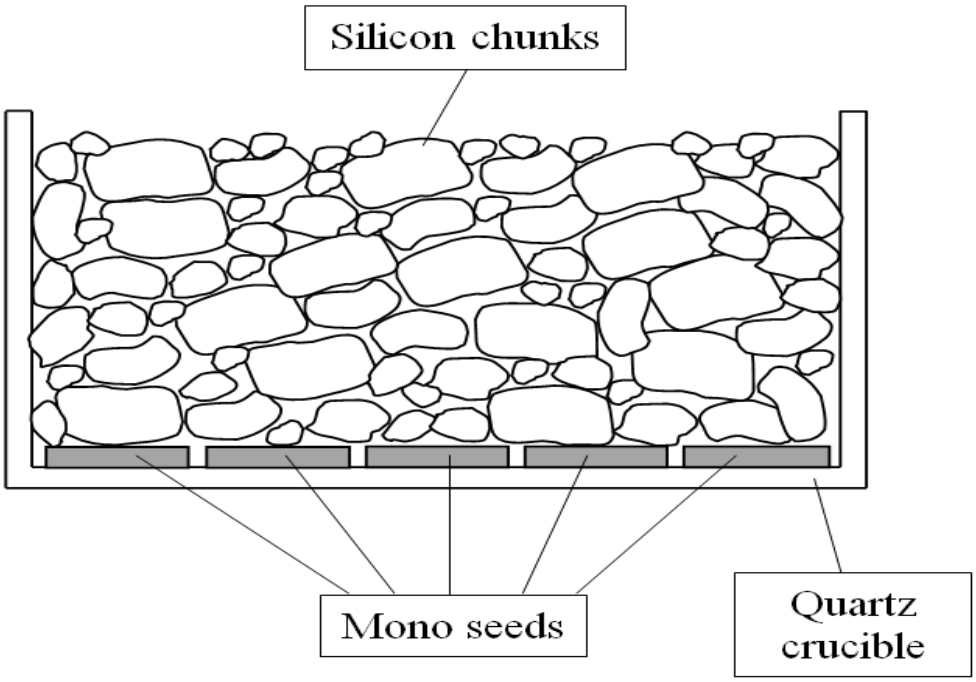


Figure 3.4: Adapted processing of DS feedstock for moonlike DS ingot production [24].

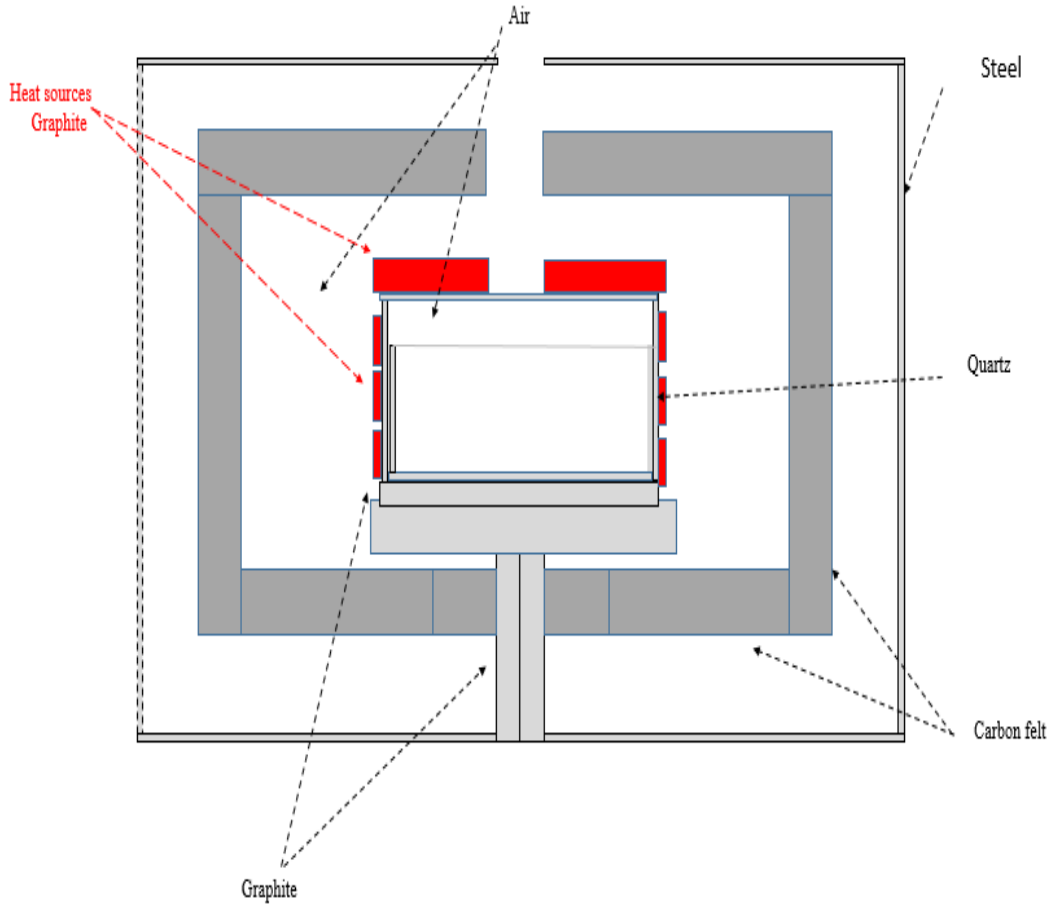


Figure 3.5: The schema of the DS furnace used for the simulation.

3.6 Description of DS Furnace Components

3.6.1 Insulation: Graphite Felt (Fibre)

The Graphite fibre is rigid graphite fibreboard, it is an insulating material made from graphite fibres and carbon binder. Based on high purity graphite fibres which have been milled and carbonized with proprietary resins, and vacuum formed into boards.

3.6.2 Insulation: Alumina Fibres

The Alumina Bulk Fibres are high-purity insulation made of polycrystalline Alumina fibres. These fibres exhibit useful properties in thermal insulation and advanced material

applications with high temperatures. They exhibit lightweight, low thermal conductivity; low thermal mass and immunity to thermal shock. Many grades of Alumina Bulk Fibres are available, making them useful in multitude applications.

3.6.3 Heating Elements (Resistor): Solid Graphite

The solid graphite has a very high density and high electrical and thermal conductor. It is a semi-metal and can be used as electrodes or resistor elements. Graphite holds the distinction of being the most stable form of carbon under standard and high temperature conditions; it is considered the highest grade of coal in the market.

3.6.4 Shell Construction: Stainless Steel

Stainless steel is a steel iron-based alloy, containing less than 1.2% of the carbon and more than 10.5% of chromium, have the property to be little susceptible to corrosion and does not degrade to rust. The stainless steel has a high resistivity form the temperature and can be used at high temperature process; with high thermal conductivity it can be used for shell structure with cooling systems.

3.6.5 Crucible: Quartz

Quartz is made of crystallized silica and present in a pure state, sometimes with a translucent aspect in rock crystal, most often opaque, or as a constituent of granite, sandstone or even sands. Pure, it can be coloured by impurities.

Table below presents the physical properties of the components of the DS furnace with the Silicon and Sapphire materials used in the model simulation:

Tableau 3.1: Physical properties of the DS furnace components, Silicon and Sapphire.

Physical Property	**Steel**	**Air**	**Carbon fibre**	**Graphite**	**Quartz**	**Silicon**	**Sapphire**
Thermal conductivity $(W/m.K)$	15	0.04	160	1790	1931	2533	27.21
Density (Kg/m^3)	7900	120	0.4	100	4	40	3970
Heat capacity $(J/Kg.K)$	477	1035	2035	1800	1060	140	419
Fusion temperature(K)						1685	2323

3.7 COMSOL Thermal Model

We used the COMSOL thermal model to simulate the evolution of the thermal field in the whole furnace. The heat is imposed by heat sources in solid graphite resistors in the furnace as shown in Figure 3.5. The furnace and its environments can be considered as a homogeneous and isotropic domain. All the components of the DS furnace can be assimilated as part of the domain of our numerical simulation system; to resolve the thermal filed configuration of this system, we can use the local equation of the heat transfer (3.2), which allows, after resolution, to determine the temperature at any point in the system at every moment.

$$\rho c \frac{\partial T}{\partial t} = div(\lambda \vec{\nabla} T) + P \tag{3.2}$$

In our case, λ can be considered constant considering our system is a homogeneous medium and λ independent of T :

$$\rho c \frac{\partial T}{\partial t} = \lambda div(\vec{\nabla} T) + P \tag{3.3}$$

$$\rho c \frac{\partial T}{\partial t} = \lambda div(\nabla^2 T) + P \tag{3.4}$$

$$\frac{\partial T}{\partial t} = \frac{\lambda}{\rho c} \nabla^2 T + \frac{P}{\rho c} \tag{3.5}$$

Where:$\nabla^2 T = \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}$ (Laplacien),.

Equation (3.5) is the heat equation that controls the thermal evolution in the furnace, Trepresents the calculated temperature (K), ρ represent the density of the medium (kg/m^3), $c$represents the Specific heat energy at constant pressure $(J/K.kg)$, P represents the thermal sources heat (W/m^2) and λ represent the thermal conductivity of each medium $(W/m.K)$.

In our case, the COMSOL model will solve equation (3.5) in all domains that represent a 2D symmetric geometry of the DS furnace by calculating the thermal conduction process in unsteady state.

3.7.1 Initial and Boundary Conditions

The initial and boundary conditions use for the simulation are as follows:

- An initial temperature which is equal to the ambient temperature $T = 289.15K$.
- The physical properties of the subdomains (subdomain by material) which are connected by numerically coupled walls.
- In the shell construction we have the circulation of water, to keep the temperature constant: In our case $T0 = 330K$.
- The heat sources P is imposed in the resistors (solid graphite elements).

3.8 Simulation Results and Discussions

We did the simulation for both materials Silicon and Sapphire and for the three following cases:

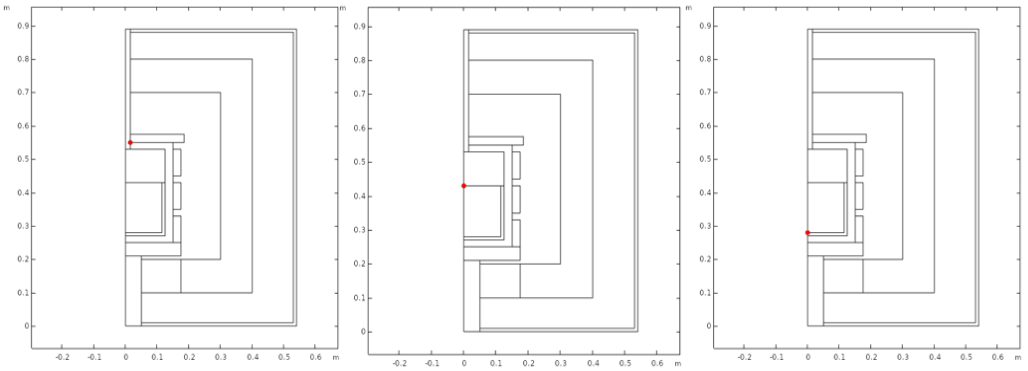
- **Case 1:** The standard furnace without modifications, top and side resistors are powered by heat sources. (Figure 3.6, Case1)
- **Case 2:** We introduced to the standard furnace in the bottom an additional resistor powered with a heat source. (Figure 3.6, Case 2)
- **Case 3:**We made some changes on the bottom material physical properties of case 2. (Figure 3.6, Case 3)

Tableau 3.2: The new physical property of the bottom material for case 3.

Physical Property	**Graphite low density instead of Carbon fibre**	**Graphite low density instead of Graphite**
Thermal conductivity$(W/m.K)$	5	5

We will present the thermal evaluation of the following parameters:

- The evaluation of the temperature as a function of time for the points (in red) which represent the thermocouple locations in the DS furnace which are represented in the following figures:



TC 1: located in the top heater **TC 2:** located in the top charge **TC 3:** located in the bottom charge

Figure 3.6: Position of the thermocouple (red points) in DS furnace for the thermal analysis.

- The evaluation of the temperature as a function of the distance in the lines (in red) which represent the top of the seeds and the bottom of the charge which are represented in the following figures:

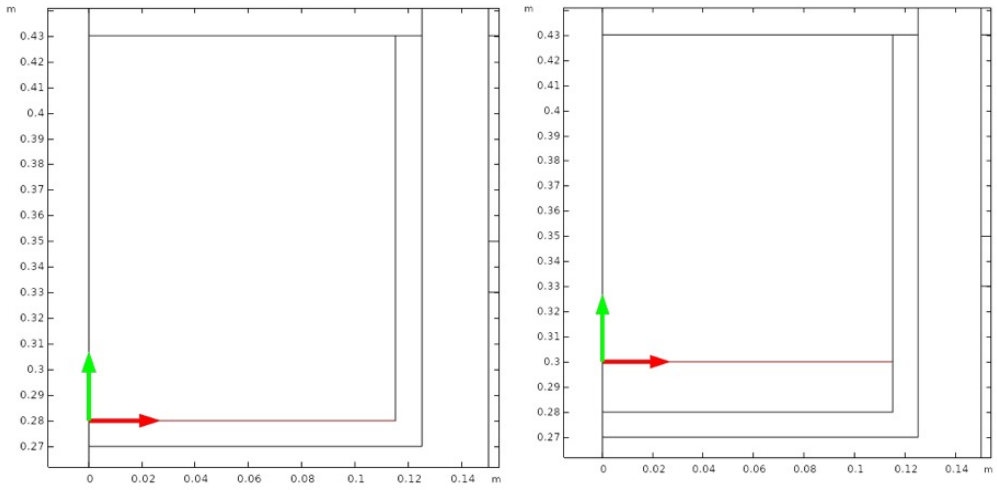


Figure 3.7: Position of the lines (in red) in the DS furnace for the thermal analysis.

3.8.1 Silicon Results: Case 1 (Standard DS Furnace)

The results in the following figures show the thermal filed and the evolution of the max temperature with time obtained for the standard furnace, Case 1:

The thermal configuration shows that the time needed to melt completely the charges is about 30 hours with a max temperature of 1886K.

To understand what is happening, the figure below shows the evolution of the temperature in the predefined lines:

We can notice two observations on this figure:

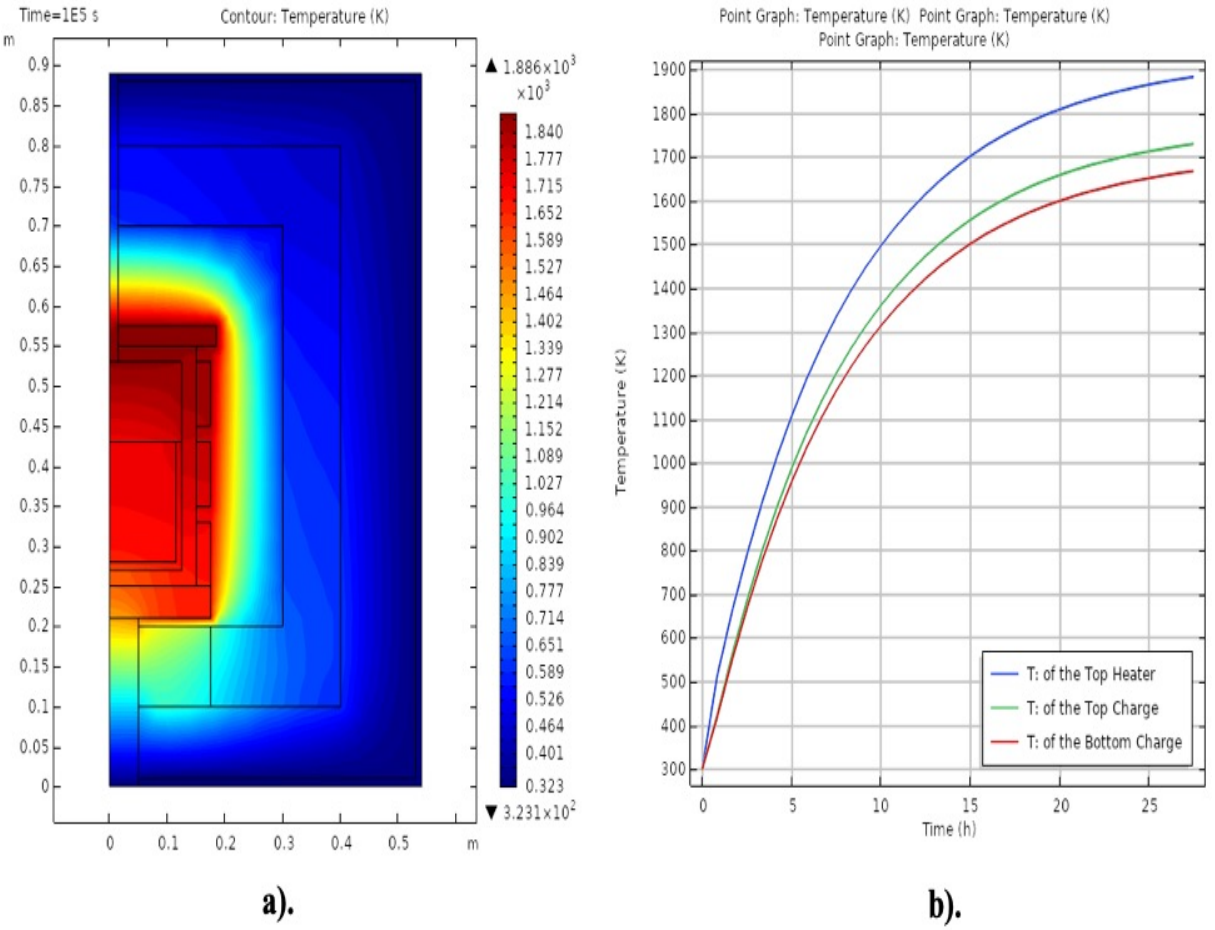


Figure 3.8: Case 1 for silicon material, (a). Thermal field configuration and (b). Thermocouples (TC1, TC2 and TC3) temperature evolution with time.

- The first one, there is a difference of 16-20 degrees between the bottom of the silicon charge and the top seeds (at 2 cm up).
- And the second, the two lines are in the same temperature range, in the middle area for the bottom charge line and on the side area for the top seed line.

This means that when we reach the melting temperature in the upper side of the seeds in the middle area, we will completely melt the seeds in the side area of the crucible. In this situation, we will lose the germs on the sides of the crucible.

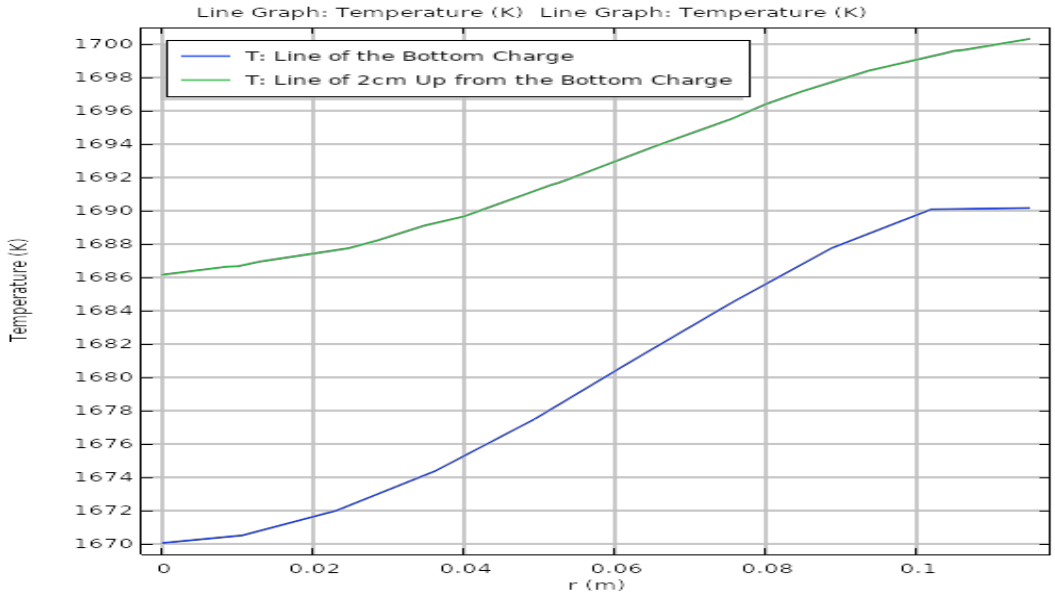


Figure 3.9: Case 1 for silicon material, the temperature values on horizontal lines 1 and 2.

3.8.2 Silicon Results: Case 2

The results in the following figures show the thermal filed and the max temperature as a function of the process time for Case 2:

The thermal configuration shows that the time needed to melt completely the charge is the same as in case 1 with a maximum temperature equal to 1830K. As case 1, the evolution of the temperature in the predefined line is presented in the figure below:

The Figure shows that we reduced the difference of the temperature between the two lines to 7-13 degree in the bottom of the Silicon charge and the top seeds (at 2 cm up), but when we reach the melting temperature in the top seeds in the middle area, we will melt completely the seeds of the side area. In this situation, we still lose the germs on the sides of the crucible.

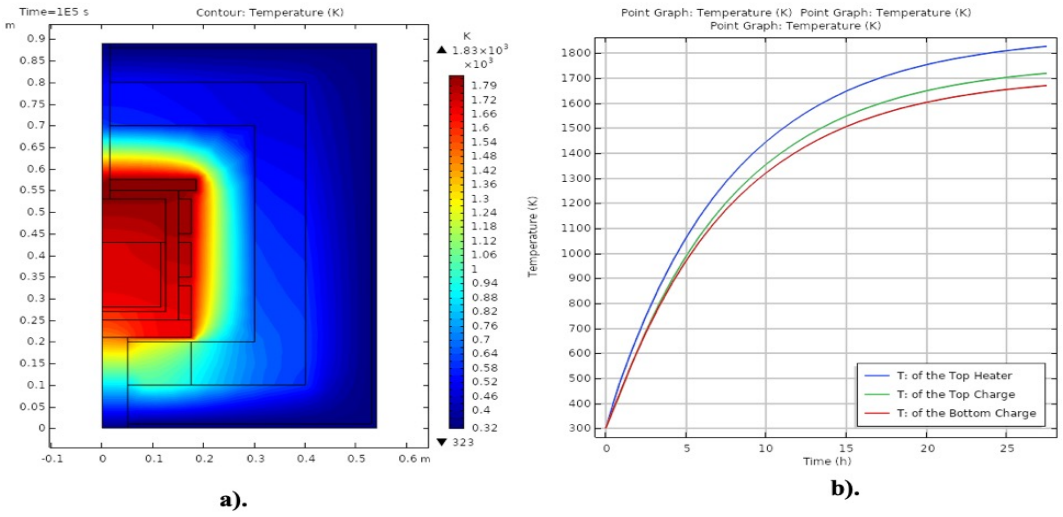


Figure 3.10: Case 2 for Silicon material, (a). Thermal field configuration and (b). Thermocouples (TC1, TC2 and TC3) temperature evolution with time.

3.8.3 Silicon Results: Case 3

The result of the thermal evolution for Case 3 shows that the time needed to melt completely the charge is the same as in cases 1 and 2 with a maximum temperature of 1731K. To confirm this observation, the figure below shows the evolution of the temperature in the predefined line:

The figure shows that we got a high improvement with this adaptation, we reduced the difference of the temperature between the two lines to 1,5-1 degree in the bottom of the Silicon charge and the top seeds (at 2 cm up). In this situation, the seeds will be intact as mono germs in all the bottom of the crucible.

3.8.4 Sapphire Results: Case 1 (Standard DS Furnace)

As the Silicon, we will use the same presentation. The results in the following figures show the thermal filed and the evolution of the maximum temperature as a function of a time obtained for the standard furnace, Case 1:

As observed with Silicon processing, the thermal configuration shows that the maximum temperature is equal to 2630K. To understand this result, the figure below shows the evolution of the temperature in the predefined lines:

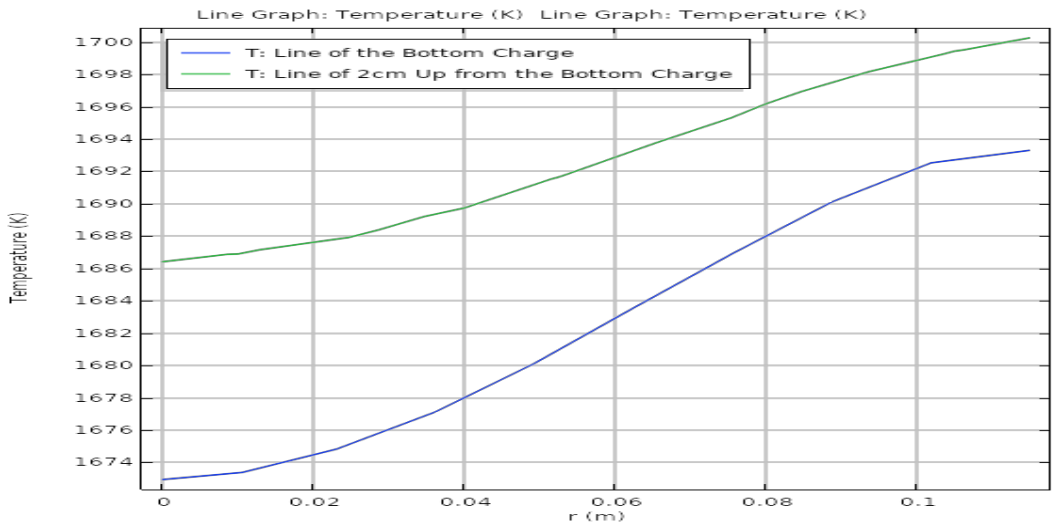


Figure 3.11: Case 2 for silicon material, the temperature values on horizontal lines 1 and 2.

We have the same two observations:

- The first one, there is a difference of 20-30 degrees between the bottom of the Sapphire charge and the top seeds (at 2 cm up).
- And the second, the two lines are in the same temperature range, in the middle area for the bottom charge line and on the side area for the top seed line.

This means that when we reach the melting temperature of the Sapphire in the upper side of the seeds in the middle area, we will completely melt the seeds in the side area of the crucible. In this situation, we will lose the germs on the sides of the crucible.

3.8.5 Sapphire Results: Case 2

After the addition of the bottom heater, the maximum temperature is equal to 2552K. As case 1, we present the evolution of the temperature in the predefined line in the figure below:

We can see that the difference of the temperature between the two lines is reduced to 20-10 degree in the bottom area of the Sapphire charge and the top seeds area (at 2 cm

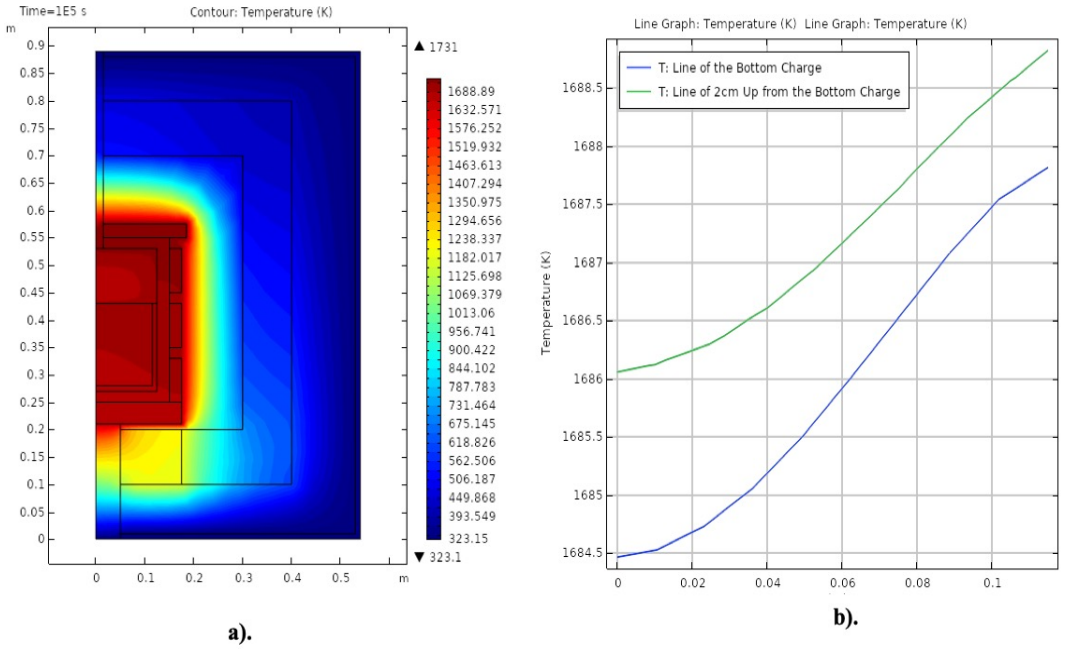


Figure 3.12: Case 3 for silicon material: a). Thermal field configuration and b). Temperature values on horizontal lines 1 and 2.

up), but when we reach the melting temperature in the top seeds in the middle area, we will melt the seeds in the sides area. In this situation, we still lose the germ on the sides of the crucible.

3.8.6 Sapphire Results: Case 3

Results show that the maximum temperature is equal to 2355K. To confirm this observation, the evolution of the temperature in the predefined line is presented in the following figure:

As the Silicon, the Sapphire processing can be improved by the new additions. The figure shows that we got a high improvement, we reduced the difference of the temperature between the two lines to 2,5-1 degree in the bottom of the Sapphire charge and the top seeds (at 2 cm up).

In this situation, the seeds will be intact as mono germs in all the bottom of the

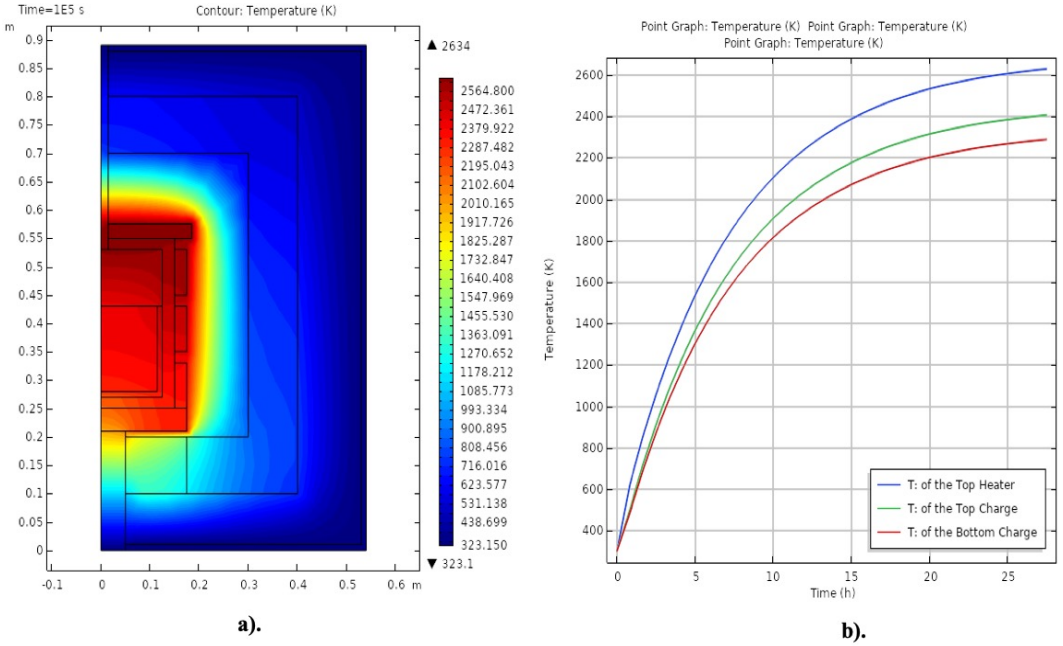


Figure 3.13: Case 1 for Sapphire material, (a). Thermal field configuration and (b). Thermocouples (TC1, TC2 and TC3) temperature evolution with time.

crucible.

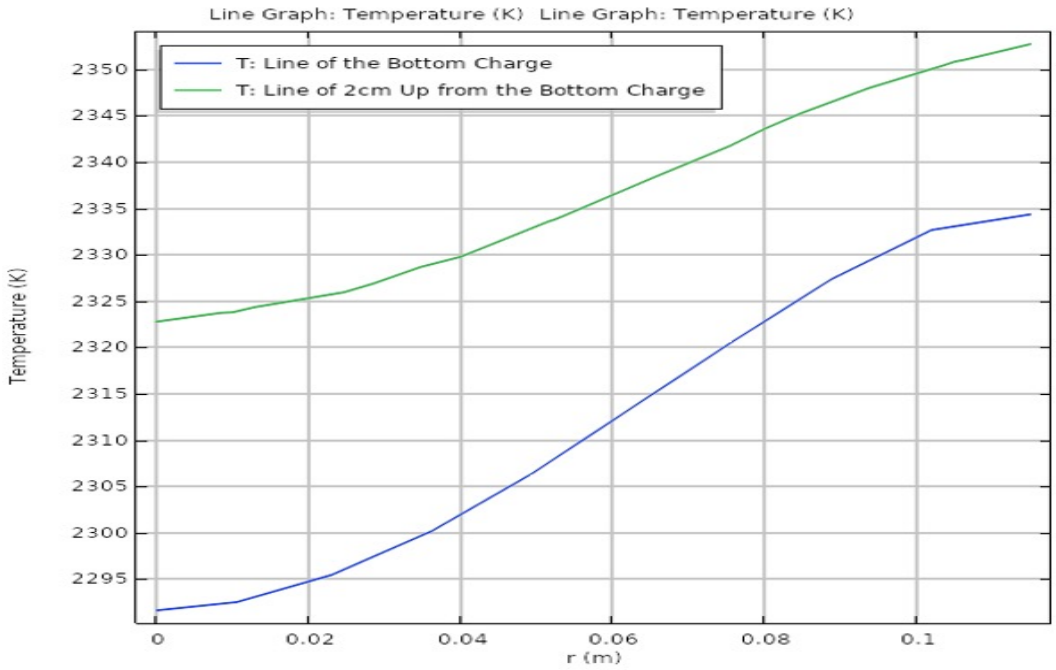


Figure 3.14: Case 1 for Sapphire material: Temperature values on horizontal lines 1 and 2.Case 1 for Sapphire material, (a). Thermal field configuration and (b). Thermocouples (TC1, TC2 and TC3) temperature evolution with time.

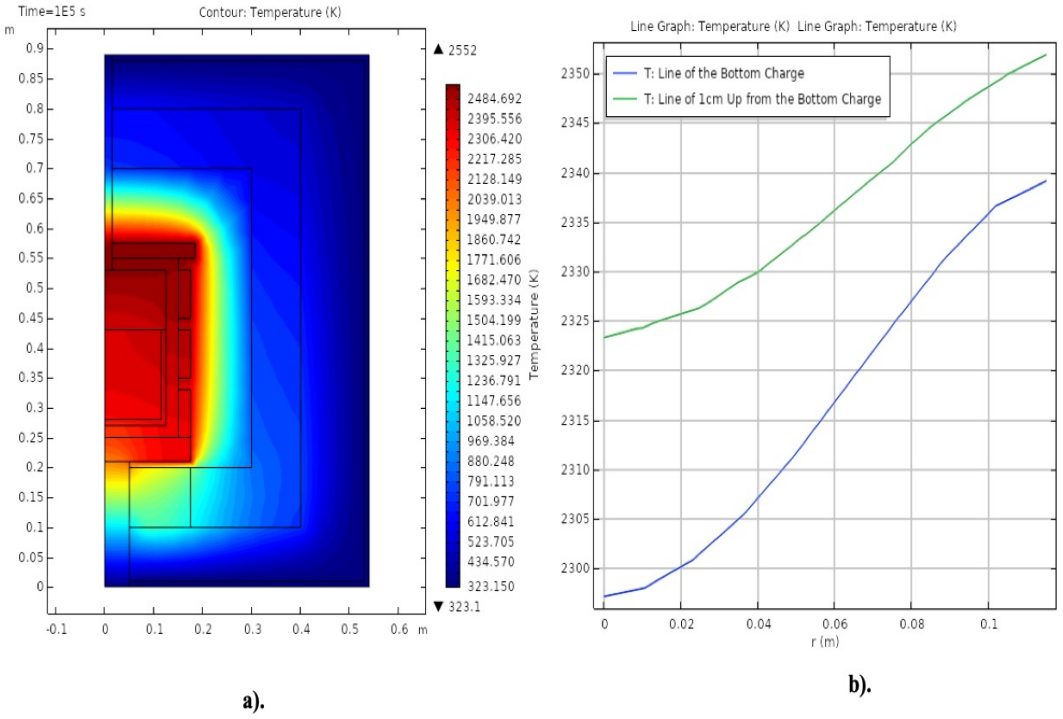


Figure 3.15: Case 2 for Sapphire material: (a). Thermal field configuration and (b). Temperature values on horizontal lines 1 and 2.

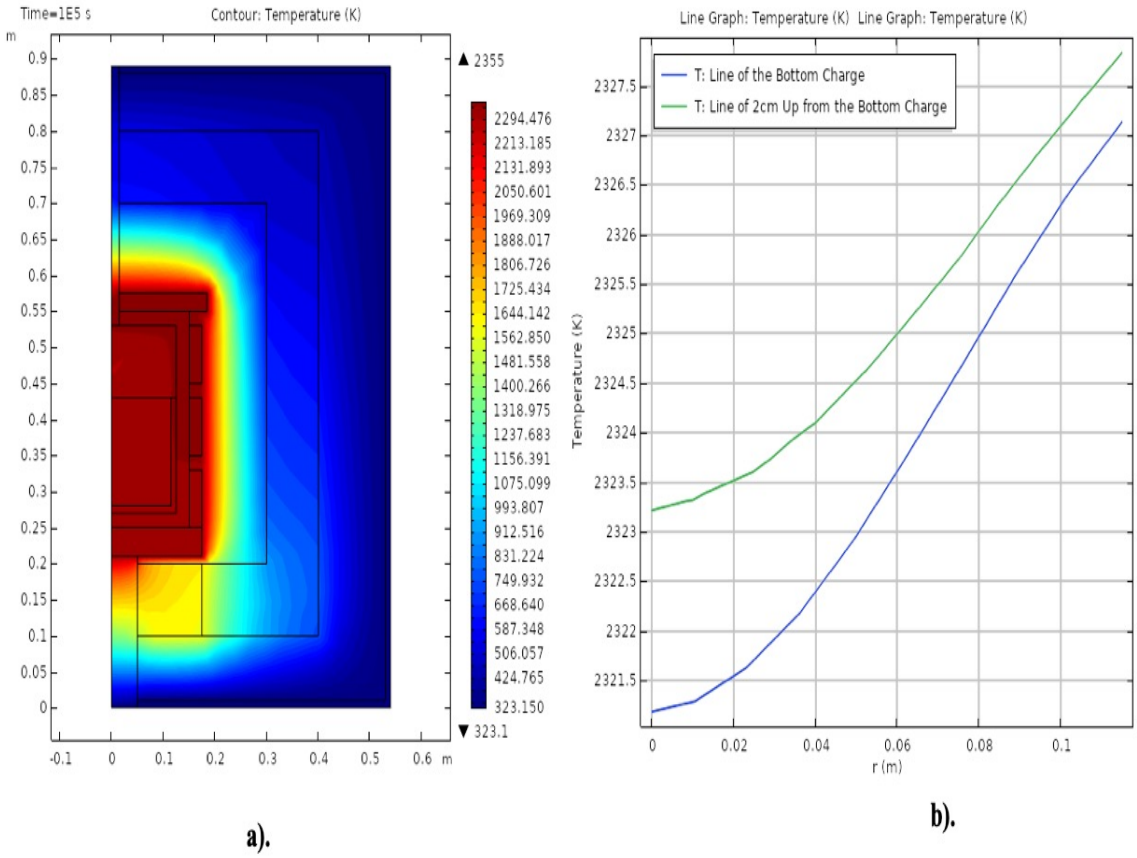


Figure 3.16: Case 3 for Sapphire material: (a). Thermal field configuration and (b). Temperature values on horizontal lines 1 and 2.

General Conclusion

The work undertaken in this study made it possible to propose a new adaptation for the DS furnace, which is widely used in materials processing, to produce a high quality of Silicon and Sapphire ingots cheaply as monolike crystal.

For this, in the first chapters we give a brief description of the important elements which are related to the raw materials and the processing methods for the Silicon and Sapphire production, and in the last chapter, we were able to carry out a very interesting numerical simulation with multi-physics software, to clearly identify the problematic and provide some solutions.

The numerical study made it possible to understand the DS process and suggest some adaptions to the standard DS furnace; we made two steps of adaptations, the first one with adding a heating element on the bottom side of the crucible and the second one adding some change in the material used in the design of the furnace bottom.

The results showed that the first control made some changes in the results but not suitable for the moonlike production. On the other hand, the second adaptation is more interesting, with both changes the difference in temperature between the middle and the sides of the crucible is not high and we can melt the top of the seeds without reaching the bottom of the crucible and losing the mono-crystal germs.

With these improvements, it is possible to produce a high quality of moonlike ingots.

In perspective, it will be interesting to simulate the process by controlling the parameters that influence the solid / liquid interface curve, because the shape of the interface can have a significant effect on the quality of the final product.

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Appendix A

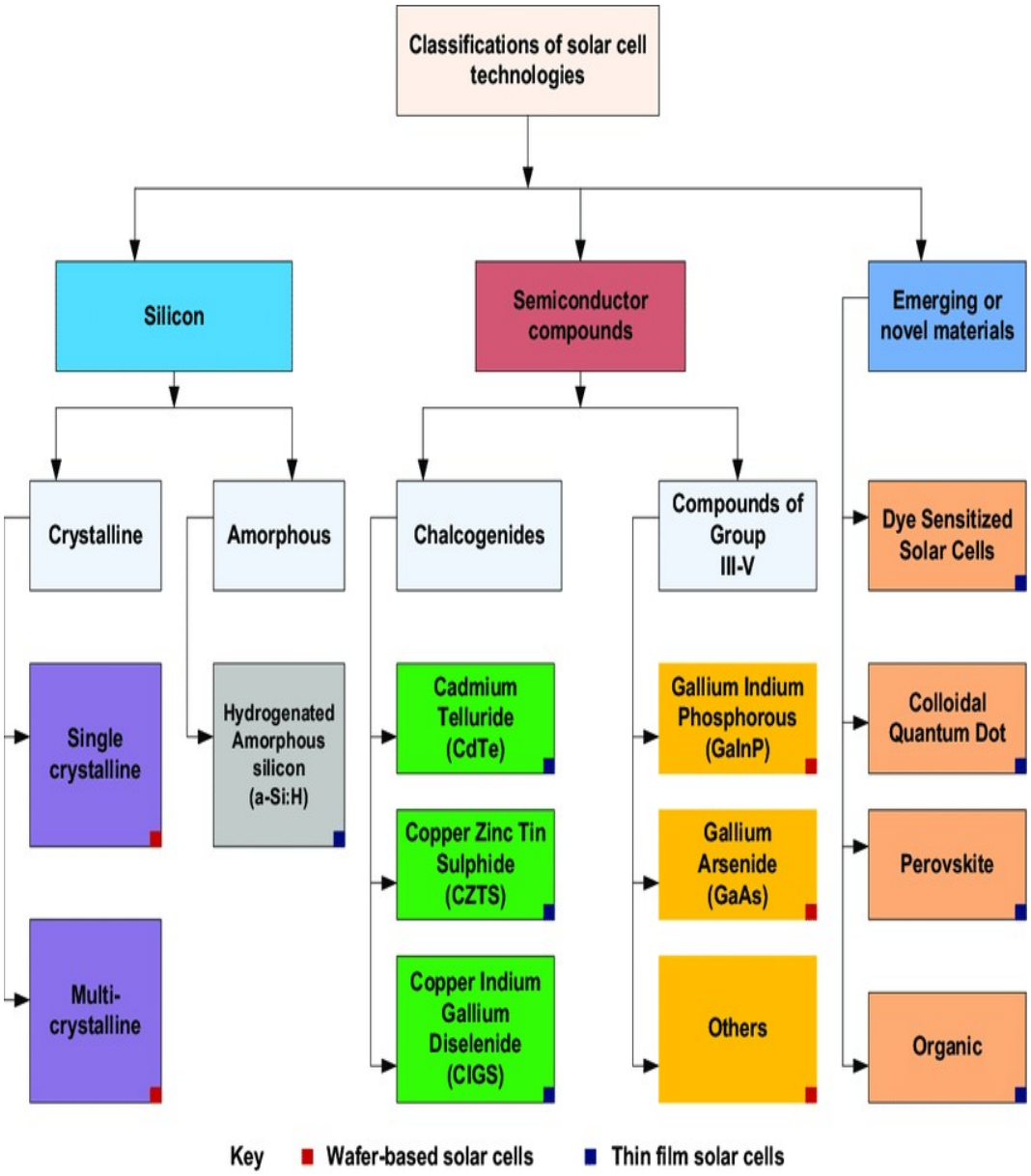

Figure 17: Classification of solar cells by technology (NREL).

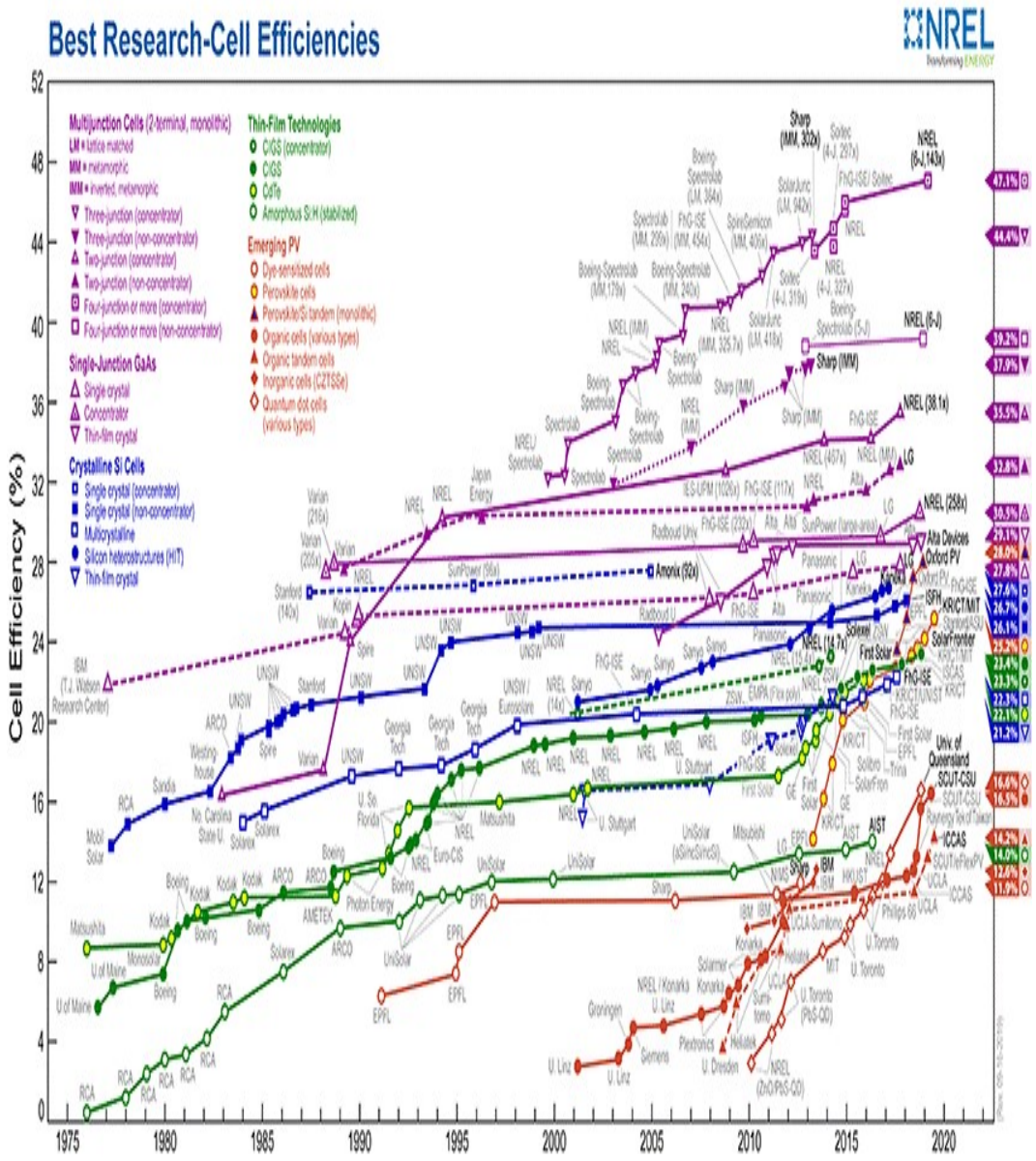

Figure 18: Confirmed PV cells conversion efficiencies for various technologies (NREL).

Appendix B

The crystallographic structure of Sapphire material is diamond crystal structure:

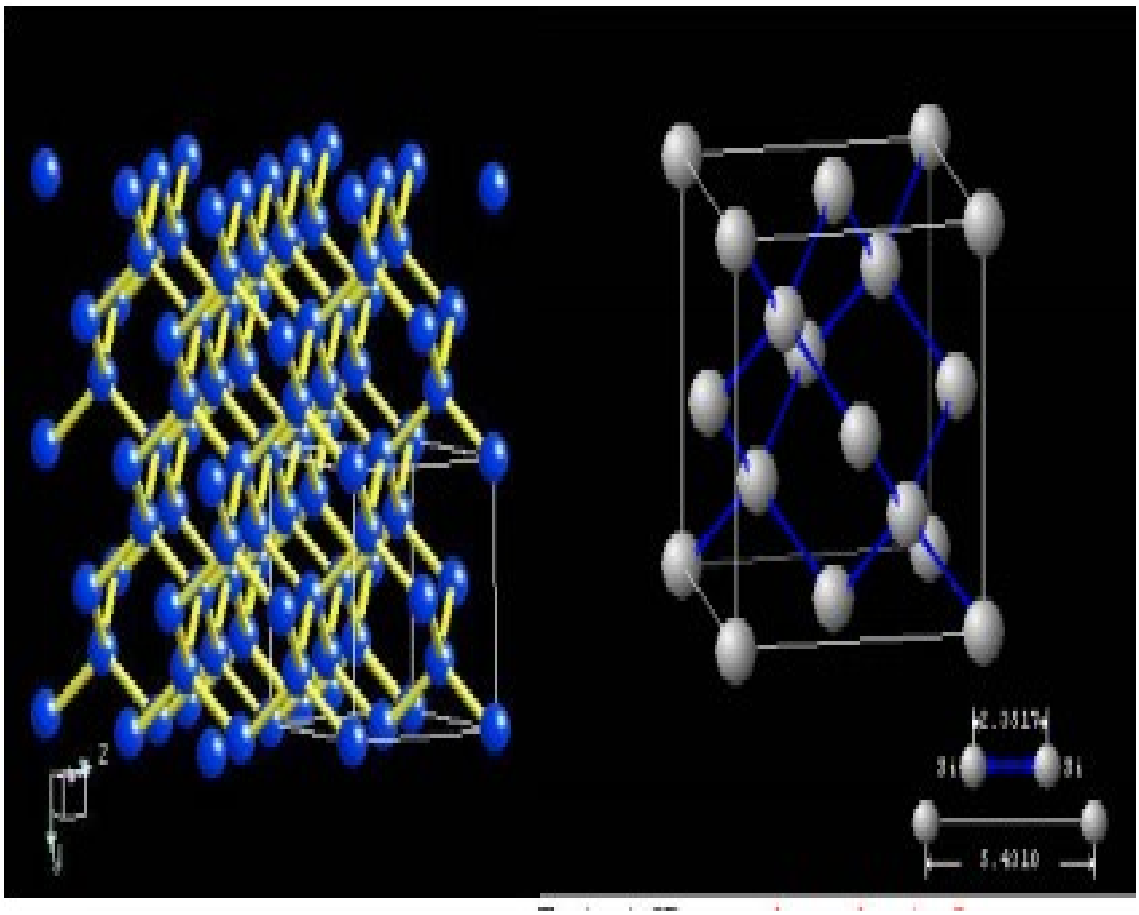

Figure 19: Crystal Structure of Silicon: Diamond Structure. From: Semiconductor Applet Service, http://jas2.eng.buffalo.edu/applets/education/solid/unitCell/home.html).

The crystallographic structure of Sapphire material is belongs to the system general trigonal with a rhombohedral hexagonal mesh, the mesh parameters are: $a = 4.76A$ and $c = 13.00A$.

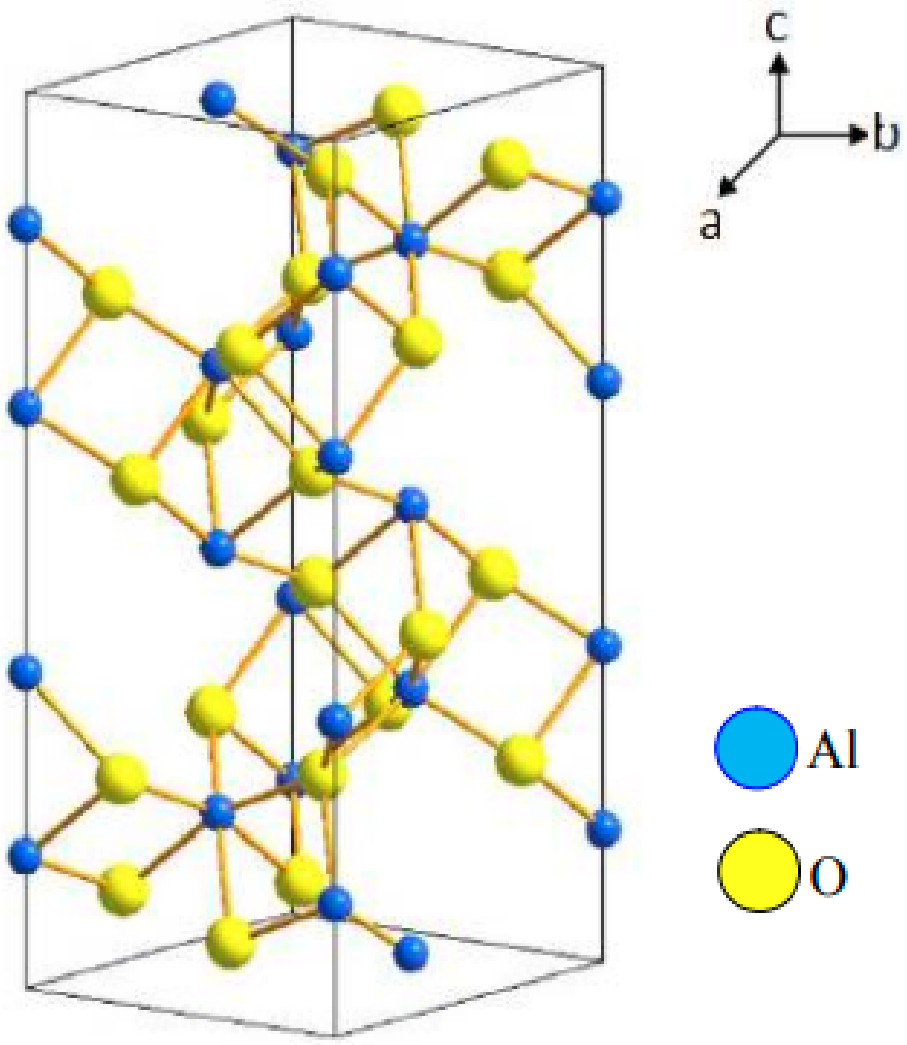

Figure 20: Structure and Elementary Cell of Al_2O_3

ملخص :

يرتبط تقليل استهلاك الطاقة في صناعة السيلكون و الياقوت بتطوير مواد و تقنيات انتاج جديدة, و كجزء من عملنا قمنا بوصف التقنيات المستخدمة في انتاج سبائك السيليكون و الياقوت بالاضافة الى تاثير عوامل التحكم الخاصة بهم ,مثل توزع درجة الحرارة في افران التصلب الاتجاهي ,قمنا باستخدام محاكاة عددية و تم اقتراح تحسينات على اساس التصميم الفعلي لفرن التصلب الاتجاهي ,من خلال هذه الدراسة تحصلنا على نتائج مهمة جدا في امكانية انتاج سبائك شبه احادية من السيليكون و الياقوت باستخدام تقنيات متعددة البلورات مع التحكم في التكوين الحراري للفرن , يمكن ان تساعد هذه التحسينات منتجي السيليكون و الياقوت على زيادة الانتاجية و تقليل استهلاك الطاقة و التكاليف.

الكلمات المفتاحية : التصلب الاتجاهي, السيليكون ,الياقوت , بذور

Abstract:

Reducing the energy consumption in the Silicon and Sapphire industry is related to the development of new processes and materials, as part of this study, we described the techniques used in the production of Silicon and Sapphire ingots as well as the influence of their control parameters, such as the temperature distribution in the directional solidification furnaces. We used a numerical simulation; improvements were made based on the furnaces standard design. The results were very interesting; they give the possibility of producing mono-like ingots of Silicon and Sapphire using multi crystalline techniques with controlling the thermal configuration of the furnace. These improvements can help the Silicon and Sapphire producers for increasing the productivity and reducing the energy consumption and costs.

Keywords : Directional Solidification, Seeds, Silicon, Sapphire.

Résumé :

La réduction de la consommation d'énergie dans l'industrie du silicium et du saphir est liée au développement de procédés et de nouveaux matériaux, dans le cadre de cette étude, nous avons décrit les techniques utilisées dans la production de lingots de silicium et de saphir ainsi que l'influence de leurs paramètres de contrôle, comme la distribution de température dans les fours de solidification directionnelle. Nous avons utilisé une simulation numérique; des améliorations ont été apportées sur la base de la conception standard des fours. Les résultats ont été très intéressants; ils donnent la possibilité de produire des lingots mono-like de silicium et de saphir en utilisant des techniques des multi-cristallins, avec le contrôle de la configuration thermique du four. Ces améliorations peuvent aider les producteurs de silicium et de saphir à augmenter la productivité et à réduire la consommation d'énergie et les coûts.

Mots-clés : Solidification directionnelle, Germes, Silicium, Saphir