



## Radyasyon Fiziği Çalışmalarında Bazı Kristal ve Amorf Malzemelerin Kullanılması

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### Anahtar Kelimeler

Katı hal fiziği  
Amorf  
Kristalografi  
Dozimetri  
Örgü  
Termoluminesans Dozimetri  
(TLD)

**Özet:** Bu derleme makalesinde, bazı amorf ve kristaller hakkında yapılan çalışmalar gösterilmiştir. Kristalografinin alanına giren, kristal örgü ve yapı konusu da, katı hal fiziği perspektifi altında açıklanmıştır. Kristal büyüme yöntemleri de kısaca açıklanıp, obsidyenin termoluminesans dozimetri (TLD) için kullanılması tavsiye edilmiştir. Obsidyen, radyasyon korunması üzerine yapılan çalışmalarda kullanılmıştır. Bu makalede, obsidyenin radyasyon dedeksiyonu için uygun bir malzeme olabileceği düşünülmüştür.

## Usage of Some Crystals and Amorphous Materials in Radiation Physics Studies

### Keywords

Solid State Physics  
Amorphous  
Crystallography  
Dosimetry  
Lattice  
Thermoluminescence Dosimetry  
(TLD)

**Abstract:** In this review article, studies of some amorphous and crystalline have been viewed. Lattice of crystalline related with crystallography has been elucidated on the perspective of solid state physics. Furthermore, usage of obsidian has been recommended for thermoluminescence dosimetry (TLD). Crystalline growth methods have been explained briefly. Obsidian has been utilized in studies which are related with radiation protection. In the article, obsidian has been contemplated as a suitable material for radiation detection.

### 1. Introduction

Main purpose in this review article is to introduce studies about some crystalline and amorphous materials related with radiation physics applications and also another aim is to propound a new idea with respect to amorphous material such as obsidian whether can be used in radiation applications or not. Dosimetric application of both nuclear and radiation physics have made a huge stride on development of new dosimeters which are utterly crucial to understand the nature of some crystalline and amorphous materials. Material at higher temperatures can emit red (infrared-visible light) radiation and materials which will be used in dosimetric studies would be either semiconductors or insulators. Metals will not be appropriate tool to observe a luminescent properties. Luminescence effect has been appeared by heating materials which are suitable as mentioned above.

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Heating is a kind of radiation and when the heat has stimulated, the material will store some energy with ionizing radiation, which means material absorbs some energy. Then, stored energy has been released as visible light but if the material has been cooled, TL properties (Thermoluminescence) of material have been terminated. Re-exposed process of ionising radiation is necessary to see visible light upon crystal or semiconductor again [1].

OSL (Optically Stimulated Dosimetry) is a procedure or natural material such as quartz or feldspar [2]. Because of that reason, optically stimulated dosimetry has been suggested by scientists [3]. Radiation dosimetry generally has been used in the application such as health physics, x-ray studies, radiation protection and so on. Borate phosphors is effective and close to human tissue with an atomic number which is  $Z=7.4$ . Because of that reason, coefficient of absorption of borate phosphors is as same as biological materials. Borate phosphors has some advantages which are as follows; wide energy band gap, sensitivity of neutron, thermal stability, optical characteristic and etc [4], [5]. Piezoelectric quartz has been studied in dosimetric application as dosimeter. In a study which focuses on development of quartz dosimeter and determining of different corrosivity caused by photooxidizing agents and volatile organic acids has been resulted by using reactive lead coating was applied to quartz crystal. Quartz vibrates coating lead that has an effect on lower frequency [6]. The other study on energy band gap of potassium lithium borate glass has been carried out by the applying FTIR (Fourier Transform Infra Red) and UV-VIS (Ultraviolet-Visible Spectrophotometry) spectroscopy techniques [7]. Next study has been related with the thermoluminescence of C doped lanthanum aluminate crystalline ( $\text{LaAlO}_3:\text{C}$ ) synthesized by **Solid State Reaction** with U.V (Ultraviolet) dosimetry [8]. Another research has been done about the subject which is thermoluminescence and radioluminescence of a sodium doped feldspar. Sodium has been used for hole traps (Correcher et al, 2007) [9].

One of the oldest studies in dosimetry of alkali halide crystal has been irradiated with gamma and x-rays [10]. Photoconductivity has been efficient on imperfection lattice of crystals by doping H (hydrogen) or F (fluorine) [11], [12]. Another study in luminescence about crystalline has been done by scientists (Jang et al, 2012) on  $\text{Eu}^{+3}$  doped fluorthalenite ( $\text{Y}_3\text{Si}_3\text{O}_{10}\text{F}$ ) for laser excitation spectroscopy for understanding temporal behavior of crystal [13]. Next research has been done on Ag doped and undoped lithium tetraborate ( $\text{Li}_2\text{B}_4\text{O}_7$ ) by using **Czocharlski Method** (Kurali et al, 2016) [5]. Furthermore, diamond dosimeter has been studied and these types of dosimeters are adequate devices for small field dosimetry. Single crystal devices have shown efficient performance, where polycrystalline can be into bigger size and they also have demonstrated low quality due to structural imperfection [14].

Another study on radiophotoluminescent dosimetry for carbon and magnesium doped  $\text{Al}_2\text{O}_3$  crystal [15].  $\text{Sm}:\text{CaNb}_2\text{O}_6$  single crystal has been studied by Czocharlski method which includes high x-ray diffraction, optical absorption and fluorescence [16]. Thermoluminescence dosimetry properties of Ge doped photonic crystal fibres (PCFs) have been evaluated with **Modified Chemical Vapour Deposition** method (MCVD) by scientists [17]. LiF material has been doped with elements which are P, Mg and Cu in a study to grasp the effect of thermoluminescence [18]. Hydrogenated amorphous silicon has been studied due to its own intrinsic radiation hardness for dosimetry and particle detection such as x-rays. Hydrogenated amorphous silicon has been thought as efficient candidate of radiation resistance quality [19], [20]. TLD property of pencil lead graphite has been evaluated in another scientific study [21]. Another investigation have been done  $\text{K}_2\text{YF}_5:\text{Tb}^{+3}$  as well as undoped  $\text{K}_2\text{YF}_5$  has been synthesized under hydrothermal conditions. Linearity of dose response and reproductivity of dose have been emerged by scientists [22]. Lucin metavariscite ( $\text{AlPO}_4 \cdot 2\text{H}_2\text{O}$ ) has been studied with different characterization techniques. TL has an important role to see structural changing in material under  $150^\circ\text{C}$ - $165^\circ\text{C}$ . Transformation of metavariscite into berlinite has caused by loss of water molecules and crystal lattice changing from monoclinic to trigonal [23]. Kinetic parameters of La doped phosphate glass under the TL glow curves of beta radiation by applying some methods such as peak shape (PS) methods and computerized glow curve deconvolution code (CGCD) [24]. In another paper, magnesium oxide (MgO) with co-doped  $\text{Li}^+$ ,  $\text{Ce}^{+3}$  has been synthesized with the method called **Solid State Reaction Technique** (Guckan et al, 2021) [25]. Other research has been done on boron doped hydrogenated amorphous silicon by using co-sputtering technique related with PECVD film samples [26]. In another research, amorphous silicon sensor has been put on the top of integrated circuit and radiation detectors based on amorphous silicon industrialized in very thin films and also amorphous silicon has been used in solar cells for space application [27], [28], [29]. A new study on susceptibility of obsidian has been done in Japan and also obsidian has been characterized with different crystals such as microlites and phenocrysts [30]. Goksu and Turetken had studied on natural sources such as obsidian [31]. Other research is about to obsidian pellets in a powder will be adding teflon and these pellets have been exposed to gamma radiation of  $^{60}\text{Co}$  in dose interval between 10 Gy-10 kGy and TL, OSL, TSEE techniques have been used in study of obsidian materials which can be a candidate of detector [32]. Alzahrani et al, investigated on obsidians under U.V. the situation of TL and PTTL [33].

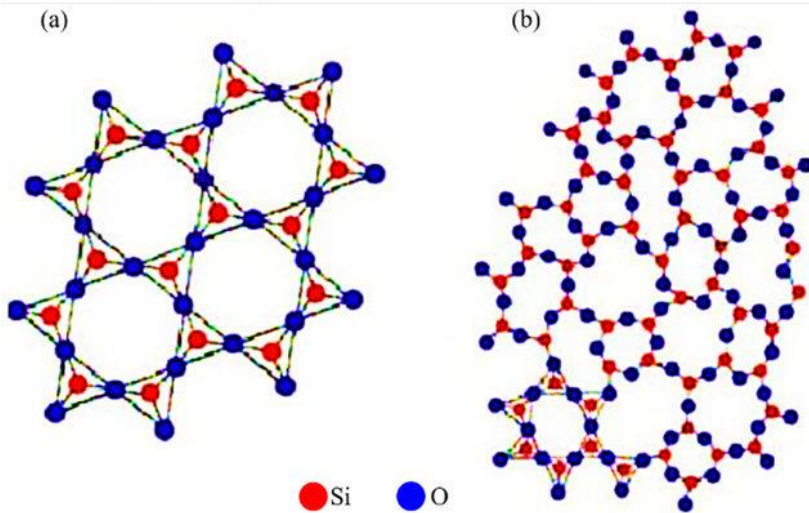
The writer has written an article related with obsidians and he has made a mistake by writing a sentence “% 75 of obsidian contains quartz” which has not been proved in any study. These both materials are similar to each other chemically because of  $\text{SiO}_2$  compound and furthermore, the writer of this article had thought that obsidian can be useful for radiation detection and it can be an adequate tool as target for particle accelerators. He has declared an opinion about this subject upon his own previous article [12].

## 2. Basic Information about Amorphous and Crystal

In the beginning of the 1900's, development of quantum mechanics had a crucial impact on forming the theory related with the energy band gap of solids. Correlation between crystals can be managed with the mechanic, thermal, electrical and magnetic qualities of solids. The main aim of crystal physics is to understand relationships between particles of microscopic and crystal structure [34]. Crystal physics studies consist of geometrical forms and physical properties such as doing experiment upon x-rays, electron, neutron beams and etc. Johannes Kepler had thought on the reason why snowflake has 6 corners instead of has 5 or 7 corners. He had made assumptions on the geometry of crystalline. Robert Hooke and Rene Just Haüy had studied on providing the mathematical theories[37]. Picture 1 shows amorphous and crystal stones which are taken from general chemistry notes[35]. Picture 2 demonstrates lattice models of both amorphous and crystal solid[36].



**Figure 1.** Galena( $\text{PbS}$ ), Pyrite( $\text{FeS}_2$ ) and Quartz have crystal structure and obsidian is an amorphous material, (General Chemistry Notes)



**Figure 2.**Crystal and amorphous lattice ( Atif et al,2020)

Amorphous means “ shapeless” and that word originally comes from ancient Greek word which is “ámorphos” [35]. Crystal lattices are consisted of three dimensional system. These systems are originated by atoms or group of atoms. In three dimensional space, according to position of atoms in crystal there are 14 different lattices or Bravais lattice and the word crystal comes from “krystallos” which means “ice” [37].

### 2.1. Classification of Crystal Lattices

Cubic system: In this type of lattice system there are three different models which are; primitive, body-centered and face-centered. Connection of any two different cubic lattices is impossible. Tetragonal system: This type has included primitive and centered and there is no Bravais connection. Orthorhombic system: in this system there are four types of lattice that can be categorized as; primitive, base centered, body centered. There is Bravais connection between primitive and base centered which is defined as “phase transition” [38]. Trigonal or Rhombohedral: It has three equal planes and two angles are also equal except one angle. Hexagonal system: It has six rotational axis and every each corner has one atom. One atom has placed in the surface and in the center, there are three atoms. Monoclinic system: It has different sides and one angle is not equal to  $90^\circ$ . Triclinic system has no equal sides and angles are not equal to  $90^\circ$ . Table 1, can give an information to the readers with different properties of crystals with respect to their geometric shapes [39]. Coordinate system have been defined as three dimensions and have been composed of non-coplanar vectors. Basis vectors can be called as  $a, b$  and  $c$ . Angle of intervectors  $\alpha, \beta, \theta$  are called as metric parameters. Different evaluations of basis have been originated by scalar products of all basis vectors. Changing from one basis to another basis has been explained by transformation matrix ( $P$ ). This transformation can be described as in the followings;

$$(a, b, c)P = (a', b', c') \quad (1)$$

$$G' = P^T G P \quad (2)$$

$G$  and  $G'$  stands for metric tensors [40].

**Table1.** Types of lattice in crystals (Kashif,2018).

Systems	Axis/Sides	Angles	Lattice Symbol	Examples
<b>Cubic</b>	a=b=c	$\alpha=\beta=\theta=90$	P,I,F	Fe,Cu,NaBr,Au,Diamond,NaCl
<b>Hexagonal</b>	a=b $\neq$ c	$\alpha=\beta=90, \theta=120$	P	Mg,Zn,Graphite,,ZnO,Ice
<b>Tetragonal</b>	a=b $\neq$ c	$\alpha=\beta=\theta=90$	P,I	NH <sub>4</sub> Br, SnO <sub>2</sub> , Sn
<b>Rhombohedral</b>	a=b=c	a=b=c<120( $\neq$ 90)	P	Bi, NaNO <sub>3</sub> , KNO <sub>3</sub> , As, Bs
<b>Orthorhombic</b>	a $\neq$ b $\neq$ c	$\alpha=\beta=\theta=90$	P,I,F,C	BaSO <sub>4</sub> , K <sub>2</sub> SO <sub>4</sub> ,
<b>Monoclinic</b>	a $\neq$ b $\neq$ c	$\alpha=\theta=90\neq\beta$	P,C	Sugar, Sulfur
<b>Triclinic</b>	a $\neq$ b $\neq$ c	$\alpha\neq\beta\neq\theta$	P	K <sub>2</sub> Cr <sub>2</sub> O <sub>7</sub> , H <sub>3</sub> BO <sub>3</sub>

## 2.2. Amorphous Materials

Macroscopic crystals are made of microscopic sized crystals. Positions of every each atom in spectacular crystal have been extrapolated by the formula as following;

$$r_{atom} = M(r_0, atom) + u.\vec{a} + v.\vec{b} + w.\vec{c} \quad [41], [42]. \quad (3)$$

Positions of atom in amorphous materials have not been calculated as positions of atom in crystals. Amorphous materials have been supposed as disordered crystalline solids [43]. Most of amorphous solids do not shape up into crystalline form. Amorphousness is generally explained by what it is not rather than what it is in lattice of solid. Three methods have been used to determine the identity of amorphous solids in which has been carried out for experiment related with solid-state physics.(i) calorimetry to calculate temperature transition of glass (ii) x-ray scattering (iii) NMR to comprehend correlation of bonds in solid. Problem of disordered crystalline can be repaired but a new method should have been considered to grasp the structure of random atom in amorphous material. Set of random data has been set up with theoretical point of view. The meaning of "disordered" is generally described in dictionaries as unpredictable and something opposite to the order [41].Studies in amorphous liquids have been accelerated with theoretical approach by Bernal[44] . In the last two decades, study interests have been divided into two different streams which are; (i) studies related with packing of both spheres and molecules [45], (ii) simulations for atoms by **ab-initio method**[46]. Simulation modelling has been applied in the packing of spheres randomly with extremely cold conditions such as the rate of the order  $10^{15}$  K/s. Even so these methods succeeded to simulate the structure of amorphous materials by considering atomic arrangements which possessed imperfections. Ideal amorphous structure has not been officially accepted and lattice of atomic arrangements in amorphous materials have been keeping this mystery [47]. Adams and Matheson had studied on computation of dense random packings of hard spheres[48]. ,Finney also has done scientific study on random packings and the structure of simple liquids[49]. Bennet studied on defining of packing of spheres for creation of ideal amorphous solids. Bennett started with regular tetrahedron and formed random packing. It has been proved precisely which one has imperfection by Clarke and Jonnson [50], [51.]There are different scientists that made a precious contribution on the subject of packing spheres either related with lattice or not related with lattice [52], [53], [54]. Obsidian is the natural amorphous material which can solidify when it interact with oxygen. Amorphous solids can be categorized into two main groups;

- Crystallisable (metallic and metalloid glasses)
- Naturally non-crystallisable (cross- link polymers, atactic organic polymers) [41] .

When ab-initio has been emerged with Hamiltonian equation that let us to conceive intuitional with perspective of quantum mechanics

$$I\theta(t) = \exp\left(-\frac{iHt}{h}\right)I\theta(0) \quad (4)$$

This equation managed interaction between atoms and also carried out the tendency of self assembly and furthermore, fluctuations took over the dynamic of the system. Random Gaussian distribution has been followed by spontaneous density fluctuations [55], to put it simply, all materials comprise larger or lesser defects which refer to crystalline structure, or contain flaws which correspond to amorphous materials. Ideal amorphous solid model is to represent the spheres/atoms which are randomly packed and non-intersection spheres and there are two rules for the perfectly random arrangements of atoms;

- There are three non-touching spheres and centers of these three spheres which are adjacent should create irregular triangles [56].
- Spheres should be in fixed position for mono atomic amorphous solid which is ideal in fixed position when  $4 \leq k \leq 9$  touching contacts with spheres which are neighbours, for  $k > 9$  sphere is in fixed position permanently [57].

Amorphous solid which is obsidian can be heated into quartz tube at the temperature in the range between 100-700°C. TL measurements were determined by Harshaw 3500 TLD apparatus with light pulses. First TLD read out confirmed to use the time from the room temperature which goes linearly to 400°C and second readout belongs to black body radiation[33]. Black obsidian has been crushed into small pieces and then it has been pulverized by agate mortar and pestle porcelain and powder has been weighed, washed and dried for 24 hours. It was mixed with teflon by using liquid nitrogen to confirm homogeneity. Obsidian+ teflon have been irradiated at the center of radiation technology, IPEN, Atomic Energy of Canada LTD with Co-60. All test which are done related with dosimetric properties of material with respect to TL, OSL and TSEE dosimetry. Obsidian can be a good candidate as dosimeter[64].

### 3. Methods of Crystal Growth and Usage of Amorphous in Radiation Studies

Crystal growth from liquid can be categorized into six groups[58];

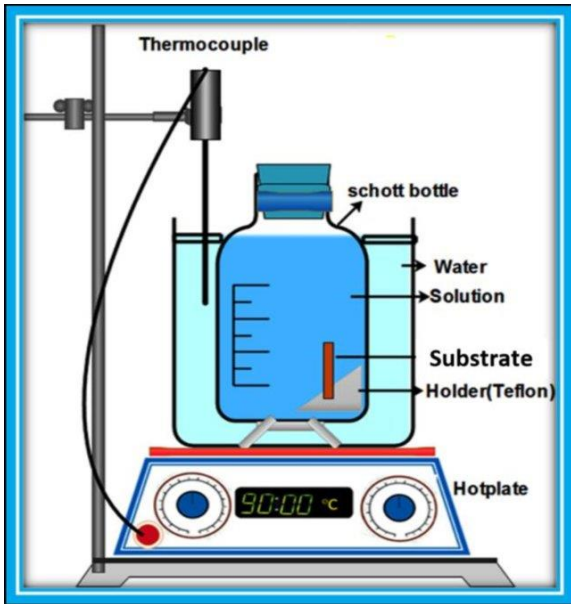
- Gel growth
- High temperature
- Hydrothermal growth
- Electrocrystallization
- Low temperature solution
- Melt growth

Alves et al, have done a research on lanthanum aluminate polycrystals by using solid state reaction method which has been mixed as equimolar ratios of both lanthanum oxide and aluminium oxide. Mixed powder has been granulated in agate mortar after powder were weighted with 0.1% weighted carbon atoms. The first approach in methodology in this study, Lanthanum oxide and aluminum oxide were put into agate mortar with 0.1 weighted of graphite and this mixture has been put into sintered process in hydrogen atmosphere at 1770°C for 2 hours. The second way of the method is aluminium oxide powder has been mixed with 0.1 % carbon and has been sintered in the same conditions. The third method is that lanthanum oxide has been put into same procedure. U.V radiation has been applied the materials and captured charged carriers removed from traps by using Regaku D/Max Ultima X-ray diffractometer. Thermoluminescent analysis has been done by using Harshaw TLD-4500 reader[8].

High temperature crystal growth has been originated by the category of high temperature solutions growth. Constituents of crystallized material were dissolved in convenient solvent and crystallization occupied as a solution which has been supersaturate. Supersaturation can be advanced by solvent evaporation with respect to cooling process of solvent or transport process in which solute is confirmed to flow from hot region to cooler region. High temperature crystal growth can be categorized into two groups which are;(i) growth from single component system and (ii) multi-component system. In this method, solid has been taken as the solvent and growth have stood under melting point. This method can be useful for oxide compounds that have high melting point [58].

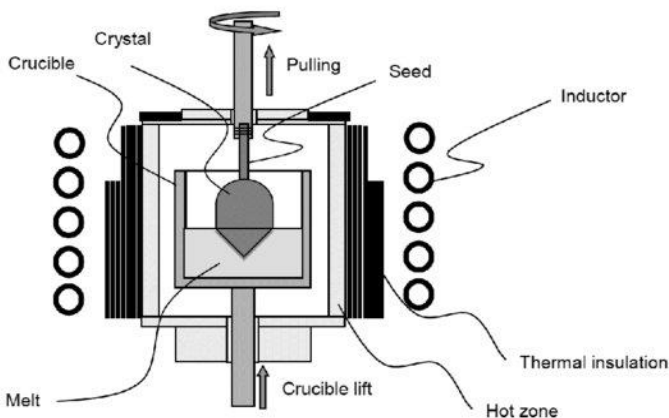
Hydrothermal growth, some materials tend to dissolve in water under high temperature and pressure in NaOH and CaCO<sub>3</sub> which are mineralization agents. Single crystal has been grown under high temperature and pressure.

Quartz can be grown by using this method which is generally called hydrothermal growth method. According to this method crystals have been grown into apparatus named autoclave and solution has placed at the bottom of hot region. Seed crystal has been placed into cooler region. Autoclave will be full of solvent which is definite amount that also can be a character as interior pressure in the system. Solution in the system will cause a rise in the heat because of temperature differences between upper and lower solution and bring solution at high temperature into growth region where material being crystallized in seed crystal[59]. Hydrothermal method has been shown in the Picture 3[65].



**Figure 3.**Hydrothermal method;( Mohammad et al, 2020)

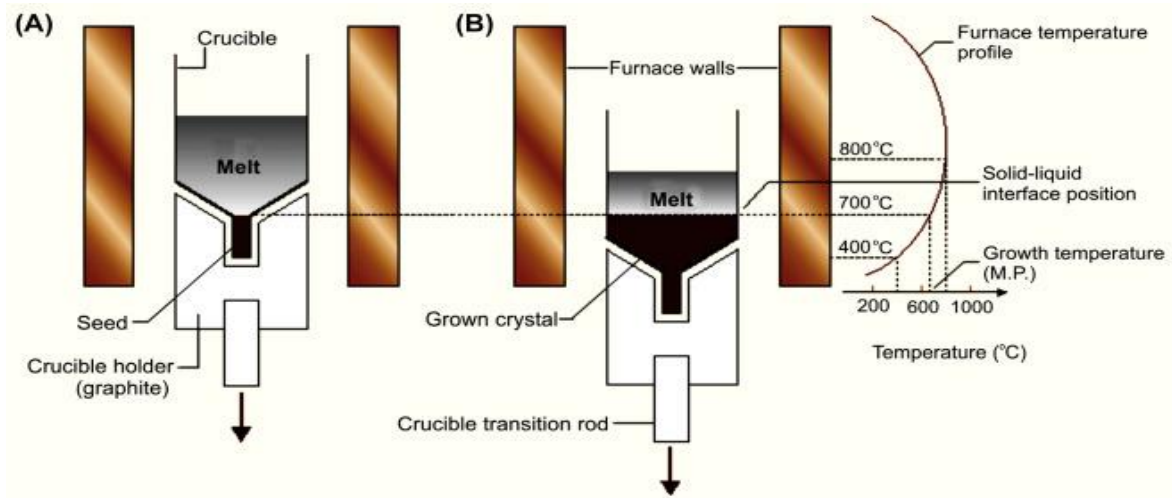
Another method on crystal growth is Czochralski method figure 4 [62]that allows obtaining crystal of many intermetallic compounds. Crystallization methods have been carried out by Czochralski in 1916 and this method focused on taking crystal out from pure metals such as Pb Zn in air[60], [61]. Then this method has been applied on the crystal growth in semiconductors and oxide for electronic industries. In this method, Crystal has been taken out from melt after seed crystal immerse into melt region and it began to grow under an adequate temperature conditions. Grown crystal has been detached from melt region when growth process has been terminated[62].



**Figure 4.** Czochralski method; (Kouwenberg,2018)

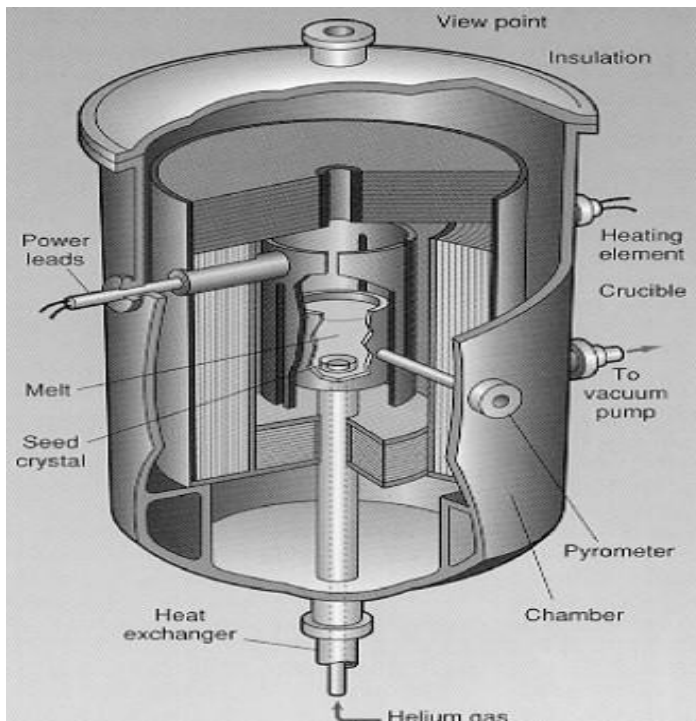
Bridgman growth method (BGO) is also used for crystal growth generally called directional solidification method. According to schematic diagram, it consists of furnace and crucible holder [63]. Seed crystal has been put into little chamber with respect to vessel and at the bottom, quartz tube has been placed vertically. Crystal solidified from the seed by changing heat element under control with increasing temperature, intersection of solid-liquid along with it will be elevated. Most of scintillation crystals have been made with this method related to the subject

of crystal growth. BGO Crystals with this method also used in satellite for discovery of dark matter in cosmology studies.



**Figure 5.** Bridgman method, (Venkataraman,2020)

Heat exchanger method (HEM) is directional solidification technique and this method includes gas-cooled heat exchanger which has been under the seed crystal. Growth has been begun by increasing cooled gas flow and decreasing temperature of furnace. Heat exchanger has a role of controlling temperature at the growth interface of crystal. Heat exchanger output has been controlled by helium gas. Sapphire crystal can be grown by HEM method. Evaporation method, circulation method, sublimation method, evaporation method, top seeded solution method, cold crucible method, floating zone method also can be used for the process of crystal growth [59]. Picture 6 has been cited from a study of Khattak and Schmid [66].



**Figure 6.** Heat exchanger method (Khattak and Schmid, 2001).

Understanding of phase equilibrium is utterly necessary knowledge to provide efficient crystal growth. Homogeneous part of macroscopic mass system shows the same atomic arrangement which is phase of material and each phase uniform structure along its volume. Phases have been ruled by Gibbs which gives a knowledge about equilibrium state of a material also it can be described as following[59];



$$F = C - P + 2 \quad (5)$$

F: number of intensive degree of free state.

C: number of components in the system.

P: number of phases.

Number 2 demonstrated both pressure and temperature which can be changed.

### Vapor Crystal System

Equilibrium temperature is  $T_0$  and saturation vapor pressure can be shown with  $P_0$ . If  $P_1 > P_0$ , vapor will be in supersaturated state and  $P_1$  stands for supersaturated vapor pressure. If vapor is assumed as ideal gas, metastable phase defined by  $P_1$  and change in free energy (Gibbs free energy) should be; [59]

$$\Delta G = \int_{P_0}^{P_1} RT_0 \frac{dP}{P} = RT_0 \ln \frac{P_1}{P_0} \quad (6)$$

(Ye, et al,2017)

R: constant of gas

$\Delta G =$  is force for crystal growth from vapor, released energy along phase transition

### Solution Crystal System

Solution and crystal in the system saturation concentration will be  $C_0$ . Constant pressure and temperature  $C_1$  bigger than  $C_0$ . Solution is supersaturated in metastable state. Driving force in this system related crystal growth will be Gibbs energy for growth process of material [59].

$$\Delta G = RT_0 \ln \frac{C_1}{C_0} \quad (7)$$

### Melt Crystal System

Molten phase and solid crystal phase endures in an equilibrium state [59];

$$\Delta T = T_m - T \quad (8)$$

$$\Delta H(T_m) = T_m \Delta S(T_m) \quad (9)$$

$$\Delta H(T_m) = \text{molar enthalpy}; \quad (10)$$

$$T_m \Delta S(T_m) = \text{Entropy difference between crystal and melt states} \quad (11)$$

## 4. Results

In this review article, lattices of both crystal and amorphous were explained concisely by guiding oldest studies which are also related with luminescence dosimetry properties. Theoretical approach has been made a clear shortly. Amorphous structure has been clarified by considering that it has flaw and crystal has disorder. Some hydrogenated amorphous silicon has been doped by H or F to observe photoconductivity properties. Thermoluminescence, carried out interaction between radiation sources and response of material in which goes through process of crystal growth method or mixing process (doping as PECVD method) for amorphous. Obsidian can be an efficient dosimeter for development of dosimetry and it can be useful on the subject of different studies related high energy physics such as particle accelerators for discovery of dark matter, neutrino and so on.

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