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# Preparation for Ultra High Pure Indium Metal for Optoelectronic Applications

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***Abstract—Ultra high pure Indium metal is extensively used in optoelectronic devices. Indium and its alloys become potential candidates in aerospace, defense and communication sectors. Purification of Indium has been done by Instrolec-200 Refiner followed by Directional Melting/ Freezing and Solidification Systems. Major targeted impurities are Metallic impurities Ag, Al, As, Bi, Ca, Cu, Fe, Ga, Ge, Mg, Pb, Sb, Si, Sn, and Zn. Purified Indium is characterized by analytical techniques Inductively Coupled Plasma- Optical Emission Spectrophotometry and Inductively Coupled Plasma- Mass Spectrometry.***

*Keywords*—**Indium, Purification, Instrolec-200 Refiner, Metallic impurities**

## I-INTRODUCTION:

**S**PHALERITE ores, Lead and Tin circuits, Zinc Flue Dust and Waste Liquors from Indium Plating are main resources from which Indium metal is recovered. Indium is minor metal in the earth's crust but it has unique and versatile combination of properties, Low melting point (156.5987$^{o}$C) and high boiling point (2072$^{o}$C) [5]. At -269.63 $^{o}$C Indium becomes superconductor which makes it very useful in Cryogenics applications [4]. Ultra high pure Indium is used in several applications. Purification of Indium is very foremost operation. In purification process, if the purity of starting material remains low then even by using perfect and appropriate method, Ultra high pure and device grade product is very difficult to achieve. Multi Pass vertical zone refining is employed for purification of 3N pure Indium which is followed by Directional Melting/ Freezing and Solidification System. Purification is done with respect to some targeted impurities like Ag, Al, As, Bi, Ca, Cu, Fe, GA, Ge, Mg, Pb, Sb, Si, Sn, Zn and the results are obtained.

## *CONCEPT OF ZONE REFINING AND MULTI-PASS ZONE REFINING:*

It is a logical extension of the use of freezing and crystallization in the means of purification process. For zone refining, process materials have low concentration of impurities and are in solid state. For formation of molten zone in the material, a narrow segment of material is melted. Solid portion of material becomes crystalline from the molten zone. Concentration of impurities present in solid region is different from that of remaining in liquid region. To yield the principal material in very pure form, these impurities or minor components are carried forward to one end of the column. Here, the segregation of impurities which is necessary for preparing pure material is characterized by the equilibrium distribution co-efficient(segregation co-efficient) (k) of the material. . k<1 requires for obtaining high purity. Single pass zone refining process requires more time for purification which is altered by Multi-pass Zone Refining. In Multi-Pass Zone Refining, a long solid ingot is set in relative motion which is placed with number of heating elements to provide input energy for the redistribution of solute in the ingot. Many molten zones in the sample are produced instead of single zone. After a single cycle, the impurity is concentrated in many bands which are separated by clear and identifiable zones of the pure material. The removal of impurities in stages is known as multi-pass zone refining. Factors influencing the refining process are:

a) Zone travel rate
b) Direction of zone movement
c) Zone size and length of sample
d) Temperature gradient
e) Diameter of the sample tube
f) Thermal conductivities of phases
g) Latent heat of fusion
h) Density difference between solid and liquid
i) Speed of crystal formation
j) Surface tension of the liquid
k) Tendency of the liquid to super cool

**Materials and Methodology:**

***Process-1: Instrolec-200 Refiner***

The refiner is capable of handling 500 gms. of low melting points materials [melting point up to (+400$^{o}$C) and as low as (-200$^{o}$C)]. Nine heaters and eight coolers placed alternatively in the instrument along the length of the sample tube. Setup is used to create up to nine narrow molten zones in the material. The slow and reciprocal movement of the sample tube which contains the material is through the heaters. Sample tube is made up of Teflon having 20 mm dia, 520mm length and filling height around 350 mm. It can contain up to 500 grams of the material. This sample tube is subjected to rest vertically on the drive cam wheel as that is essential to place it in appropriate position between the selected two heaters and coolers. For providing such correct position, two aluminum guides are used. These 9 radiation type electrical heaters are used to produce up to nine separate molten zones along the sample tube. Temperature providing capacity of each heater is about 400$^{o}$C at a surface of the sample tube. Eight coolers are arranged to constrain the molten zone produced by the heaters. Coolers are helpful to obtain rapid and good crystallization from the molten zones. By using laboratory chiller, non-corrosive liquid is allowed to flow with pressure of 175 KNm$^{-2}$. When the melting temperature of the sample is near to the ambient temperature, a low temp. coolant (-30$^{o}$C) is used to keep molten zones narrow so rapid crystallization occurs. The movement of the sample tube is vertical and the rate of movement is 25 mm hr$^{-1}$.A digital totaling counter on the front panel is activated when the refining cycle is completed.

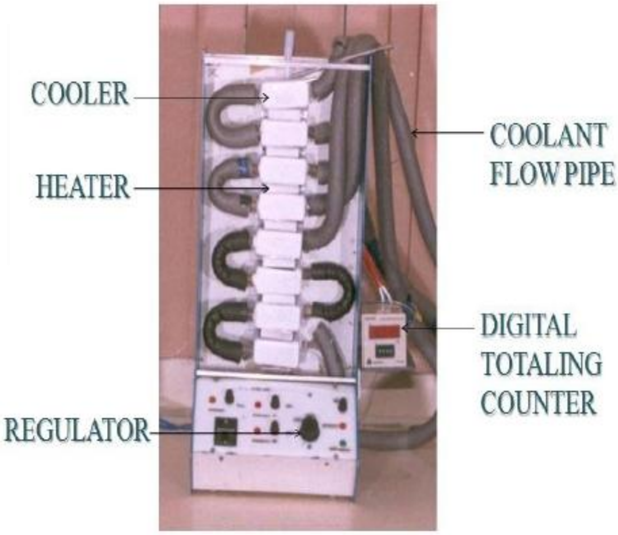


**Figure (A): INSTROLEC-200 REFINER**

***Process-2: Directional Freezing, Fabricated Proto-Type Controlled Melting And Solidification Systems***

Figure (B) shows the furnace which is used for homogenization and crystallization of the sample between these two processes. It consists of two zones, one zone is hot and rest one is gradient zone. Temp.gradient in gradient zone is about 2 to 5 $^{o}$C /cm. Temperature providing capacity is about 1000$^{o}$C. Temperature is controlled and measured by PID (Programmed Integral Differential) – Computer Controlled/Programmed. Coolant flow assembly is used to circulate coolant at optimum rate. Drive mechanism of the sample tube is operated by computer programmed.

Figure (C) shows the vertical furnace having length of 700 mm. Maximum operating temperature and continuous operating temperature of furnace is around 1000$^{o}$C and 900$^{o}$C respectively. High quality ceramic muffle alumina tube having 540 mm outer diameter is used with 2 mm wall thickness. High temperature zone length is 100 mm, low temperature zone length is 100 mm and gradient zone length is of 450 mm. Pt/Pt-Rh thermocouple is used which is controlled by compensation cable. PID controller is employed with suitable power pack. The control thermocouple which is placed close to the heating element is touching the inner quartz liner tube in the middle of the high temperature zone. Fabrication of furnace is accomplished by ceramic bricks/blocks or insulating quartz wool. For programming of furnace operation and controlling the furnace parameters, appropriate software is employed. Data logging software computer interface facility is utilized for processing furnace parameters soak, storage of data and its analysis. A vibration free up/down transverse system is made to move and rotate sample tube having 500 grams material weight with specified speed. The sample tube is held in a holder and that holder spindle is used in order to achieve transverse and rotation movement.


**Figure (B): Proto-Type /Laboratory Furnaces (750° C & 1000° C) And Sample Lowering Facility Used For Crystallization Experiments**


**Figure (C) : Computer Controlled Directional Melting/Freezing And Solidification System**

## EXPERIMENTAL DETAILS

***Process-1: Multi-Pass Vertical Zone Refining***

Granulated Indium (3N pure) of around 400 gms. was subjected to essential processes i.e.acid washing, cleaning and etching and for its purity confirmation, it is analyzed by ICP-OES

(Inductively Coupled Plasma Optical Emission Spectrophotometry). Using hot plate, sample was heated around 170°C with gentle stirring for pre-homogenization. Then the molten indium was poured into thoroughly cleaned Teflon sample tube and it was allowed to cool down to solidify the sample material. After that at pressure of 175 KN $m^{-2}$, coolant supply was turned on. For the first cycle, the drive mechanism was started and so that the sample tube is subjected to the pass through heater-cooler assembly of the refiner. The temperature control then rotated to the pre-calibrated position by rotating it in a clockwise direction. Through the stepped regulation temp. is produced from 158°C to 162°C i.e.with a gradient 4°C and with zone width of 35 mm, the solid sample which is just above the first heater allowed to melt. During the pre-stage zone refining after achieving melting, temperature was regulated to keep the molten zone as narrow as possible. Each refining cycle is of 5 hours duration. Cycle counter was set to zero and the drive mechanism started. Experiment continued until all five refining cycles are not completed. Each completed cycle was recorded on the panel counter by using a sample extractor, solidified indium was then removed

***Process-2: Homogenization, Purification, Synthesis And Characterization Of Indium Metal***

Homogenization, crystallization and synthesis experiments of indium were carried out by using vertical zone refining, directional freezing, and fabricated prototype controlled melting and solidification systems. Indium was first homogenized by using two zone furnace [Figure (B)], Indium obtained after homogenization was purified by melting and solidification processes. This homogenized and purified indium was subjected to directional freezing/crystallization for 5 pass times. To prevent contamination of indium with other gaseous impurities, the top end of the crucible was fitted with air-tight Teflon cork and it was made to travel at the rate of 2.5 cm $hr^{-1}$ after placing it in the directional solidification system. Crystallization of homogenized pure sample was done under vacuum condition in a prototype controlled melting and solidification systems and tube furnace. Final zone refined crystalline indium was subjected to analysis and characterization by ICP-MS.

## RESULTS AND DISCUSSION

To estimate the amount of specific targeted impurities such as Ag, Al, As, Bi, Ca, Cu, Fe, Ga, Ge, Mg, Pb, Sb, Si, Sn, and Zn, analytical techniques ICP-OES (Inductively Coupled Plasma Optical Emission Spectrophotometry) and ICP-MS (Inductively Coupled Plasma Mass Spectrometry) were carried out. Table-1 presents the results of purity analysis of starting indium and zone refined indium by ICP-OES and ICP-MS. The results show the significant reduction in total concentration of impurity from 93.58 ppm to 4.37 ppm in the final zone refined sample of 5N6 pure indium. Whereas the impurity concentration after step one is reduced to 13.67 ppm with respect to targeted impurities in starting 3N pure indium. Generally impurities have tendency to get reduced in refining operation. Although little bit peculiarity is shown in some impurities like Cu and Ag. After process-1, Cu and Ag impurities were not detected but after process-2, Cu and Ag were detected to 1.97 ppm and 0.87 ppm respectively. This might be possible due to experimental contamination and accidental redistribution of impurities instead of segregation.

**TABLE- 1:** Analysis of Indium

| **ELEMENT** | **STARTING MATERIAL (3N pure Indium) ICP-OES** | **After Process-1 ICP-OES** | **REFINED Indium ICP-MS** |
|---|---|---|---|
| **Ag** | 1.00 | N. D | 1.97 |
| **Al** | 2.26 | 4.45 | N. D |
| **As** | 0.05 | N. D | N. D |
| **Bi** | 3.72 | N. D | N. D |
| **Ca** | 30.00 | N. D | N.D |
| **Cu** | 3.10 | N.D | 0.87 |
| **Fe** | 5.60 | 5.75 | 0.62 |
| **Ga** | 1.74 | 1.59 | 0.81 |
| **Ge** | N. A | 0.92 | N.D |
| **Mg** | 3.02 | N.D | N.D |
| **Pb** | 3.21 | | N.D |
| **Sb** | 2.30 | N.D | N.D |
| **Si** | 28.00 | N.D | N.D |
| **Sn** | 9.58 | N.D | N.D |
| **Zn** | N. A | 0.96 | 0.10 |
| **TOTAL** | **93.58** | **13.67** | **4.37** |

## CONCLUSION:

From analytical techniques, purity of Indium is confirmed and hence we conclude that 5N6 pure Indium is obtained from 3N+ pure Indium with reference to targeted impurities Ag, Al, As, Bi, Ca, Cu, Fe, Ga, Ge, Mg, Pb, Sb, Si, Sn and Zn.

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