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Growth Techniques of Ferroelectric Single Crystals

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Abstract*— *Various growth techniques to grow ferroelectric single crystals are discussed in this paper. Some prominent methods used are mainly discussed. Some of them are Growth from the Melt which includes Bridgmann method and Czochralski method. Some of the other methods prominently used in present scenario that are discussed in this paper are Solid-state crystal growth (SSCG), Vapor-phase growth, Solid State Single Crystal Growth (SSCG), Solid-state crystal growth (SSCG), and Abnormal Grain Growth (AGG).

Keywords*— Single crystal, ferroelectric, crystal growth, techniques.

I. INTRODUCTION

Ferroelectrics: An overview

Ferroelectric materials have come up as a pathbreaking technology for a broad spectrum of semiconductor technology and electronic device applications. They find massive usage in digital information storage media and metal oxide semiconductor (CMOS) logic circuits.

Some of the areas in which ferroelectrics find a way for commercial exploitation are

i. logic and memory application
ii. organic, inorganic, and two-dimensional (2D)ferroelectric materials
iii. device structures (metal/ferroelectric/metal (MFM)
iv. metal/ferroelectric/semiconductor(MFS),
v. metal/ferroelectric/insulator/semiconductor(MFIS)
vi. metal/ferroelectric/metal/insulator/semiconductor (MFMIS))
vii. next-generation electronic devices (negative capacitance field effect transistors (NC-FETs)
viii. ferroelectric RAM (FeRAM)
ix. ferroelectric field effect transistors (FeFETs)
x. ferroelectric tunnel junctions(FTJs))[1].

A need for single crystal fabrication

Single crystals are very important group of materials becauseof their highly-ordered, continuous and uniform structure – all of which makes them possess exclusive properties. In many aspects, single crystal materials are found to have advantages over polycrystalline materials. There aremany properties in single crystals, which are not found in polycrystals [1].

In spite of the recent technological developments of advanced polycrystalline materials, which are often fabricated for specific applications, the dielectric, conductivity, electrical, optical, thermal, piezoelectric, pyroelectric, mechanical, and other properties of single crystals still remain a topic of utmost interest to explore. For this reason, single crystals and their fabrication methods are a topic of interest and concern among many scientists.

As technological development increases, there is a wide spread requirement of high-quality single crystal materials – both in bulk and in thin films.

Therefore, a huge demand of various single crystal materials has generated a necessity for improving the existing growth techniques as well as developing newer alternative techniques for single crystal growth.

II. CRYSTAL GROWTH METHODS CONVENTIALLY USED

- Horizontal Boat Growth Methods. Horizontal Gradient Freezing (HGF) method. Horizontal Bridgman (HB) method. ...

- Vertical Boat Growth Methods. Vertical Bridgman (VB) method. ...
- Pulling Methods. Czochralski (CZ) method. ...
- Floating Zone (FZ) Method.
- Other Methods. Shaped Crystal Growth Method.

Basic crystal growth methods can be separated into four categories based on what they are artificially grown from: melt, solid, vapor, and solution. Specific techniques to produce large single crystals (aka boules) include the **Czochralski process (CZ)**, Floating zone (or Zone Movement), and the Bridgman technique.

The basic growth methods available for crystal growth can be broadly classified into [1]

- Growth from melt
- Growth from vapour
- Growth from solution
- Growth from solid

2.1 Growth From The Melt

a) Bridgmann method
b) Czochralski method
c) Vernuil method
d) Zone melting method
e) Kyropoulos technique
f) Skull melting

2.2 Conventional methods of single crystal growth

Presently, there are three methods by which single crystals can be made

1) growth from melt

2) growth from solution

3) growth from vapor phase [2].

Growth from melt is the most widely used method. It is based upon the solidification and crystallization of a melted material. The Bridgman and Czochralski methods are two most used melt-growth techniques. The Czochralski method (cz) is used for the production of single crystals for many commercial exploitation.

However, the melt-growth technique shows many disadvantages in the growth process. The main difficulty lies in maintaining a constant and uniform temperature during the entire crystal growth. The challenge also lies in maintaining and sustaining very high melting points. Also achieving chemical homogeneity is a bigger task in different materials with different chemical composition .

In a material where a lot of materials are present together in the system, it poses as a tougher challenge. For making a good quality crystal, there are factors like high costs of production and equipment's, reactivity of the melted material with the crucible, chemical inhomogeneity of the input materials are also some of the factors which may contribute in making of good quality single crystal.

2.3 Czochralski method

The Czochralski (CZ) method is a crystal growth method which is widely used to grow ferroelectric single crystals. It usually initializes with insertion of a small seed crystal into a melt. The melt is often taken in a crucible. Thereafter, the seed is pulled upwards.This initiates in forming a single crystal. The method was initiated by a researcher named Jan Czochralski, who developed it in 1916.

The process starts with taking the material in a crucible. The material is then melted by resistance or radio-frequency heaters. After the material is completely melted, it is rested for some time to allow it to stabilize. Then a seed crystal is slowly inserted in the melt on the surface while giving crucible rotation and also slowly rotating the seed. A small portion of the dipped seed is melted.

Thereafter it is given some stabilizing time. It is minutely monitored to see that a melt meniscus to beinitiatedat the portion exactly where the seed and melt meet. The seed is then given small rotation time. This is all done to five a uniformly stabilizing temperature to be maintained throughout the melt.

Then, the seed is slowly pulled up from the melt (often under rotation). The growth of single crystal is usually to be seen as the seed crystal starts to swell up.The melt then starts growing at the interface by forming a new crystal. Further in the growth process, the shape of the crystal, especially the diameter, is monitored by maintaining the thermal equilibrium throughout the crucible, the pulling rate and the rotation rate of the crystal.

2.4 Solid-state crystal growth (SSCG)

Solid-state crystal growth (SSCG) has become an important technique in the development of high-quality single crystals for such systems. SSCG [4] is advantaged by its lower growth temperatures than conventional melt and solution growth techniques by producing crystals through a solid phase transformation of a polycrystalline matrix to a single crystal.

This in turn lets higher chemical homogeneity and volatility control. It also adds up to be cost-effective. Given the accurate polycrystalline microstructure and also the other processing parameters, large single crystals can be initiated. Especially those that are of incongruently melting systems those which are difficult to make using other techniques.

2.5 Vapor-phase growth

Vapor-phase growth is another method of growing single crystals. It is more pertinent to the fabrication of thin single crystals films on substrates, rather than bulk single crystals. In the instance of physical vapor transport (PVT) and chemical vapor transport (CVT) techniques, these two processes can aid in the growth of single crystals. While doing this, the vapor phase can be achieved using three processes such as reaction in the gas phase, transport reaction and sublimation process [5].

2.6 Solid State Single Crystal Growth (SSCG)

A possible way to enhance the homogeneity of crystals with complex composition is to grow them by using the low cost Solid State Single Crystal Growth (SSCG) method.

The SSCG method is essentially a form of induced abnormal grain growth, a phenomenon which is very well known in the solid state sintering community.

2.7 Abnormal Grain Growth (AGG)

A new technique of single crystal fabrication and growth, which has recently attracted lot of interest within the research community, is through the solid-state conversion of polycrystalline materials to single crystals. This technique is based on a phenomenon which can be determined in many systems, called atypical grain boom (AGG).

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