碩士學位論文

Fabrication of Rochelle Salt Thin Film

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Fabrication of Rochelle Salt Thin Film

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Contents

Abstract

Rochelle salt ($NaKC_4H_4O_6 \cdot 4H_2O$), also known as potassium sodium tartrate tetrahydrate, is a material with ferroelectricity and piezoelectricity. It had been extensively used in microphones and earpieces. Since the emerging field of low environmental impact technologies, Rochelle salt is widely investigated again. Many researches on Rochelle salt is made by cutting or combining with other materials. But Rochelle salt are made by cutting or combining with other materials is difficult to apply to manufacture a memory. In order to be used in manufacturing a low operating voltage memory, in this research fabrication of Rochelle salt crystal thin film by evaporation solution growth method is reported.

Many methods have been tried to grow crystal thin films: adjusting the size of the space, adjusting the growth direction or using different substrate materials. As a result, the length of 30.90 mm Rochelle salt single crystal is successfully grown and various thickness crystal films are fabricated in the limited space. The presence of crystallization structure films are confirmed by the presence of sharp X-ray diffraction (XRD) patterns on the samples. The sample of 2 mm thickness crystal film is single crystal and the samples of thickness less than 0.5mm (no precise measurement of thickness) crystal films are poly crystal by observing the Raman spectra. Atomic Force Microscope(AFM) is used for observing the samples surface morphology, the crystallinity of Rochelle salt on the surface of Si substrate is better than on the surface of glass is obtained. From ferroelectric measurement results, the true ferroelectric properties are not presented, but there is a possibility that Rochelle salt films can be used as a memory device is been seen.

Although the process in this work shows the possibility of producing a film with good crystal surface morphology, from experimental results: millimeter-sized single crystal film can be grown and micron-sized single crystal film growth is difficult. We need to further investigate the microscopic arrangement and orientation of Rochelle salt crystals in the future.

Chapter 1 Introduction

1.1 Research background

With the development of technology in the field of electronics, electronic devices are also developing in the direction of chip, miniaturization, thinning, low power consumption, compounding, environmental protection and energy saving. According to the report published by the United Nations University, the International Telecommunication Union and the International Solid Waste Association, humanity generated 44.7 million metric tons of electronics waste in 2016, which is equivalent to 6.1 kilograms per person [1]. Gradually, there are more and more researches on environmentally friendly and energy-saving electronic devices. Researchers have developed devices based on biodegradable materials such as polyvinyl alcohol (PVA) [2], polylactic acid (PLA) [3], or electroactive paper [4]. Also, cellulose-based electronics were studied [5].

Rochelle salt is a non-toxic and easy-to-obtain material which is one of the first-known materials to exhibit ferroelectricity [6]. Rochelle salt has two curie points, the crystal structure below 255 K and above 297 K is orthorhombic, corresponding to the paraelectric phase. While the crystal structure between the temperature 255 K and 297 K is monoclinic, corresponding to the ferroelectric phase [7].

Ferroelectric is a spontaneous electric polarization that can be reversed by the application of an external electric field. A ferroelectric material is one that undergoes a phase transition from a high-temperature phase that behaves as an ordinary dielectric (so that an applied electric field induces an electric polarization, which goes to zero when the field is removed) to a low-temperature phase that has a spontaneous polarization whose direction can be switched by an applied field [7]. The spontaneous polarization of ferroelectric materials implies a hysteresis effect which can be used as a memory function, and ferroelectric capacitors. They are also used in electromechanical transducers and actuators because the change in electric polarization is accompanied by a change in shape [7].

Under the action of alternating electric field, the polarization P of ferroelectrics changes nonlinearly with the external electric field, and in a certain temperature range, P appears as a double-valued function of electric field E, showing hysteresis. The P-E (or D-E) return line is called the hysteresis loop. The hysteresis loop can compare the maximum (saturation) polarization (Ps) of the reaction, the remanent polarization (Pr), the magnitude of the coercive field (Ec), and calculate the energy storage density of the material based on the integral of the hysteresis loop. The ferroelectric hysteresis loop is shown in Figure 1.

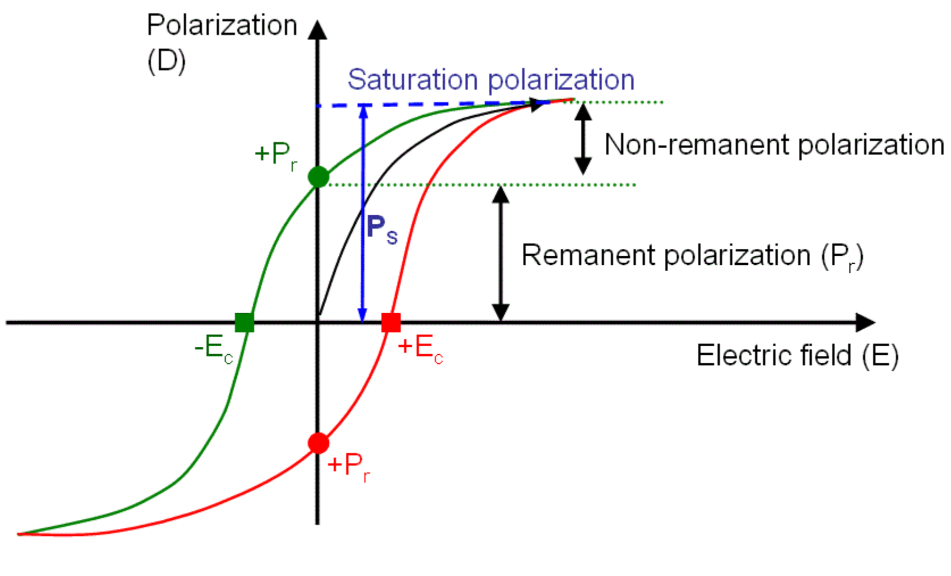


Figure 1. Ferroelectric hysteresis loop [8].

Rochelle salt also has piezoelectric. Unlike more widely-used piezoelectric materials, such as lead zirconate titanate (PZT), Rochelle salt is water soluble and does not contain toxic elements. It is an approved food additive by the Food and Drug Administration. Rochelle salt has high piezoelectric constants with a reported maximum piezoelectric tensor constant d_{14} of 2300 pC/N [9]. Rochelle salt was widely used in microphones [10] and sensors [11] during the 1940's. However, the commercial use of Rochelle salt has since been eclipsed by the development of ceramic piezoelectric materials. Since the emerging field of low environmental impact technologies, Rochelle salt is widely investigated again.

Rochelle salt crystals embedded into nanoporous materials such as glass [12], alumina [13], molecular sieves [14], or the dielectric properties of Rochelle salt incorporated into sheets of paper [15] have been studied. Rochelle salt filled polyvinyl alcohol (PVA) composite films have also been characterized [16]. However, research on micron-sized Rochelle salt crystal film is rare. For the Rochelle salt crystal film is fragile and deliquescent, the research on the application of Rochelle salt film is also a big challenge.

1.2 Motivation

Based on Rochelle salt advantages, the state of the Rochelle salt is colorless, room temperature ferroelectric behaviors and Rochelle salt crystal film growth does not require high temperature processing. Fabrication of Rochelle salt crystal film which can be applied to a low operating voltage memory was studied. Although it is very popular to study polymer materials used in memory, Rochelle salt has low coercive electric field than polymer materials. In the study of B. Fugiel et al, the Rochelle salt coercive field is about 15 kV/m

[17]. The coercive field of the polyvinylidene fluroride (PVDF) at 25℃ is 115 MV/m [18]. If combination of Rochelle salt crystal film with polymer material, whether their electronic characteristics will be changed is also worth studying.

Chapter 2 Experiment

2.1 Crystal growth

Rochelle salt is highly water soluble material and hence can be crystallized by the method of aqueous solution crystallization method. Rochelle salt crystals have been grown by evaporation solution growth method which can make single crystals without expensive equipment and be done at room temperature in this work. The Rochelle salt with a purity of 99.0% was supplied by DUKSAN in Korea and the distilled water was provided by ultra-pure water plant (Merck Milli-Q). The following is the process of crystal growth by evaporation solution method at room temperature.

2.1.1 Preparation of saturated solution

1. According to the solubility of Rochelle salt (Figure 2), determine the crystal growth temperature.

2. Weigh the corresponding amount of Rochelle salt.

3. Dissolve the Rochelle salt in distilled water.

4. If Rochelle salt does not melt well, increase the temperature to dissolve it completely (The heating temperature needs to be controlled, when the temperature is higher than 60℃, the Rochelle salt will begin to lose crystallization water). After completely dissolving, let it cool naturally to the growth temperature.

5. Filtration of solution.

6. Placed the solution under the conditions of crystal growth temperature.

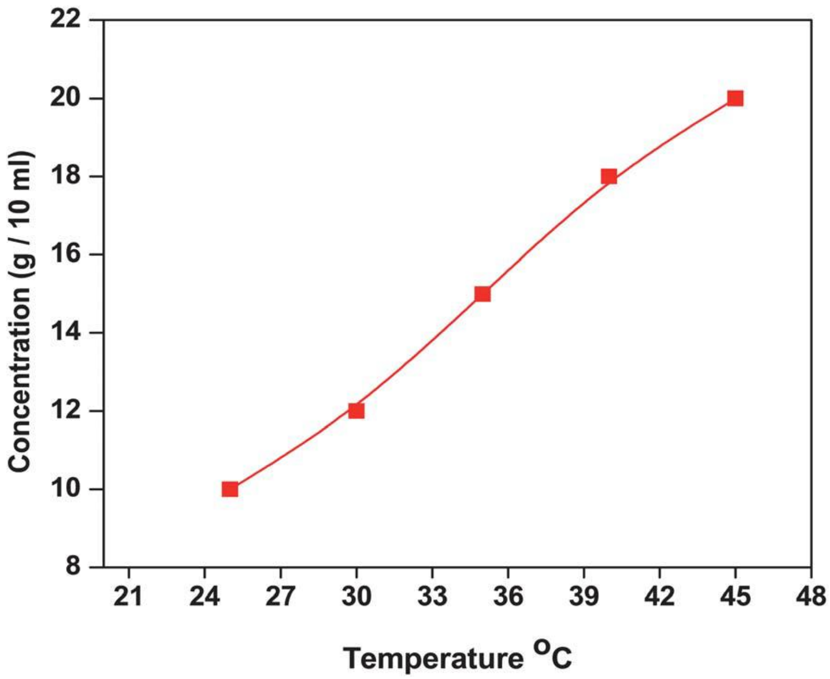


Figure 2. Solubility of Rochelle salt [19].

2.1.2 Seed crystal growth

1. Prepare clean glassware and place glassware in a place free from vibration.

2. Pour a small amount of saturated solution (20~30 ml) into the clean glassware.

3. Observe the crystal grow.

4. Select crystals with good crystal appearance, dry and store in a dry container as a seed crystal for single crystal growth (the size of the seed crystal is usually 3 to 6 mm).

2.1.3 Input seed crystal

1. Use the fishing line bind the selected seed crystal.

2. Put seed crystal to the growth solution.
3. Place the growth ampoule with solution in the thermally-isolated containe, temperature is maintained at 25℃.

4. Observe the crystal grow.

Keep all processes clean and free from vibration during crystal growth, and the temperature should be well controlled.

2.2 Fabrication Rochelle salt film

The basic idea of film fabrication is that the saturated solution is placed in a limited space and the process of crystal growth without crystal seed. The growth method is also by evaporating the solution at room temperature.

Using two parallel substrates as the growth substrates:

sample1: drop 20 drops of saturated solution via a filter paper with a diameter of 110mm on the glass substrate, and then cover at the height of 2 mm with another glass substrate.

sample2: drop 1 drop of saturated solution on the glass substrate, and then cover with another glass substrate.

sample3: drop 1 drop of saturated solution on the silicon wafer (p-type (100)), and then cover with glass substrate.

sample4: place two overlapping glass substrates vertically in the saturated solution container, therefore the edges of the glass substrates can be contacted with the solution, when the solution has evaporated, add solution (this process was repeated three times).

Growth conditions of the samples are shown in table 1, the sample growth configurations are presented in figure 3.

Table 1. Growth conditions of the samples.

sample	substrate materials	substrates direction	solution
1	glass-glass	horizontal	20 drops
2	glass-glass	horizontal	1 drop
3	glass-Si	horizontal	1 drop
4	glass-glass	vertical	unlimited

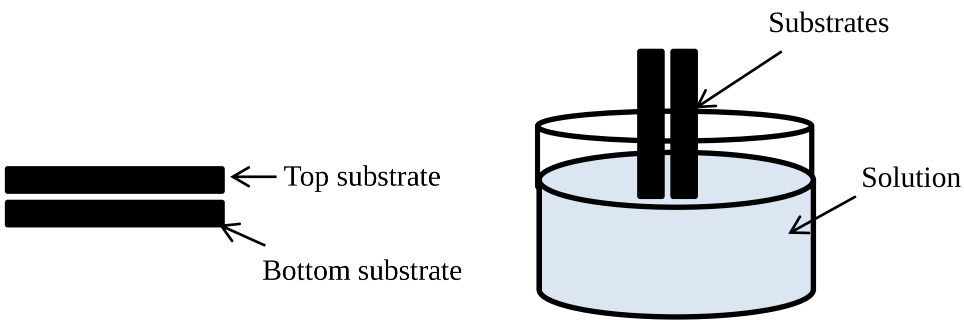


Figure 3. The sample growth configurations: horizontal (left), vertical (right).

2.3 Rochelle salt film characterization

In order to characterize the Rochelle salt film, the XRD measurement of the samples was performed by a Rigaku corporation model smart X-ray diffractometer (9kW, rotating anode), using Cu-Kα radiation.

A NOST model FEX Raman Spectrometer was used to determine the absorption bands of the samples. The measurements were carried out at arbitrary location of samples. The excitation laser wavelength was ~531 nm. The excitation laser power was ~2.2 mW, a lens of 20x (0.45NA) was used in the microscope. The spectral resolution was ~2.1-2.9/cm.

PSIA, Model XE-100 Scanning Probe Microscope was used for AFM measurement which is used to image samples surface.

Ferroelectric measurements were obtained using the RT-66A standardized ferroelectric test system.

Chapter 3 Results and discussion

3.1 Crystal growth result

After two months of growth, the length of 30.90 mm transparent columnar single crystal was successfully grown. Under suitable saturation and temperature, further growth of crystals can be seen by attracting more amounts of growth units. Figure 4 shows the Rochelle salt single crystal which was grown by evaporation solution growth method.

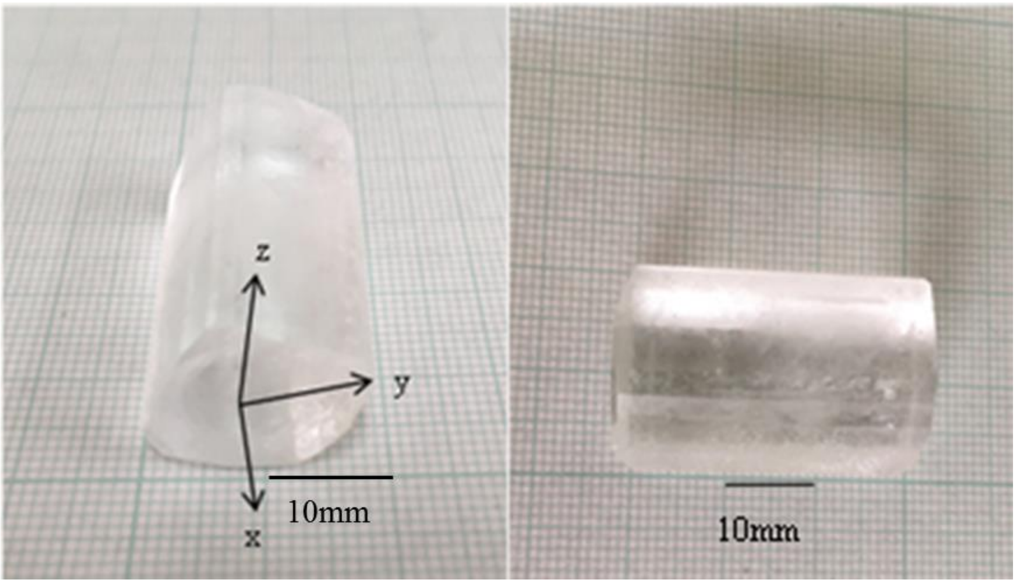


Figure 4. The Rochelle salt single crystal.

3.2 Crystal films growth result

Sample 1 grew into transparent flaky crystals after thirteen days at room temperature.

Sample 2 has a small piece crystal structure that looks like a thin film crystal in the middle of the glass substrate after five days of growth. Sample 3 grew a film similar to polycrystalline mainly in the edge portion after four days. After eight days of growth, sample 4 grew a larger area of similar polycrystalline film from the edge portion than samples that only provide 1 drop of solution. Figure 5 shows the appearance of the samples..

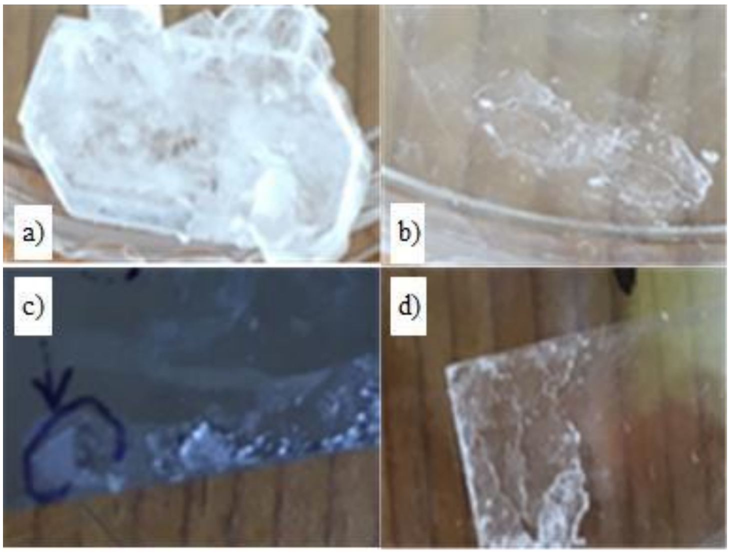


Figure 5. The appearance of a) sample 1, b) sample 2, c) sample 3, d) sample 4.

3.3 XRD measurement

In order to verify the crystallinity of Rochelle salt film, XRD measurements were carried out on the samples. Figure 6 shows the X-rays diffractograms defined Bragg peaks at specific 2θ angles. On the Rochelle salt flake crystal sample 1, three main sharp patterns at 13.02°, 34.3° and 25.54° in 2θ were recorded (Figure 6a). On the sample 2 and sample 4 which Rochelle salt film incorporated glass substrate, sharp patterns were also detected at different locations (Figure 6b, 6d). Sample 3, Rochelle salt film incorporated Si wafer, sharp patterns mainly at 13.31° and 12.42° (Figure 6c). Comparison with the XRD pattern of p-type silicon, the most intense XRD patterns of silicon at approximately 28°, 47° in 2θ [20, 21]. This means that the XRD patterns of silicon has no effect on Rochelle salt film XRD patterns. As mentioned previously, the Rochelle salt has two crystal structures: a monoclinic (between 255 K and 297 K) and an orthorhombic crystal structure (below 255 K and above 297 K). By learning and comparing relevant literatures [19, 22-24], with these XRD measurements, it is difficult to distinguish these two structures. However, the 2θ position of some of the sharp patterns detected on samples indicated that the Rochelle salt films are crystallized. The four most intense XRD peaks of the samples were list in Table 2. Whether the crystal films are single crystalline or polyrystalline will be discussed in the following Raman measurement results.

Table 2. Four most intense XRD peaks of the samples.

Sample / 2θ(°)	1	2	3	4
1st peak	13.02	12.40	13.31	44.38
2nd peak	34.3	24.94	12.42	11.92
3rd peak	25.54	11.88	24.96	13.00
4th peak	35.34	12.96	32.96	12.40

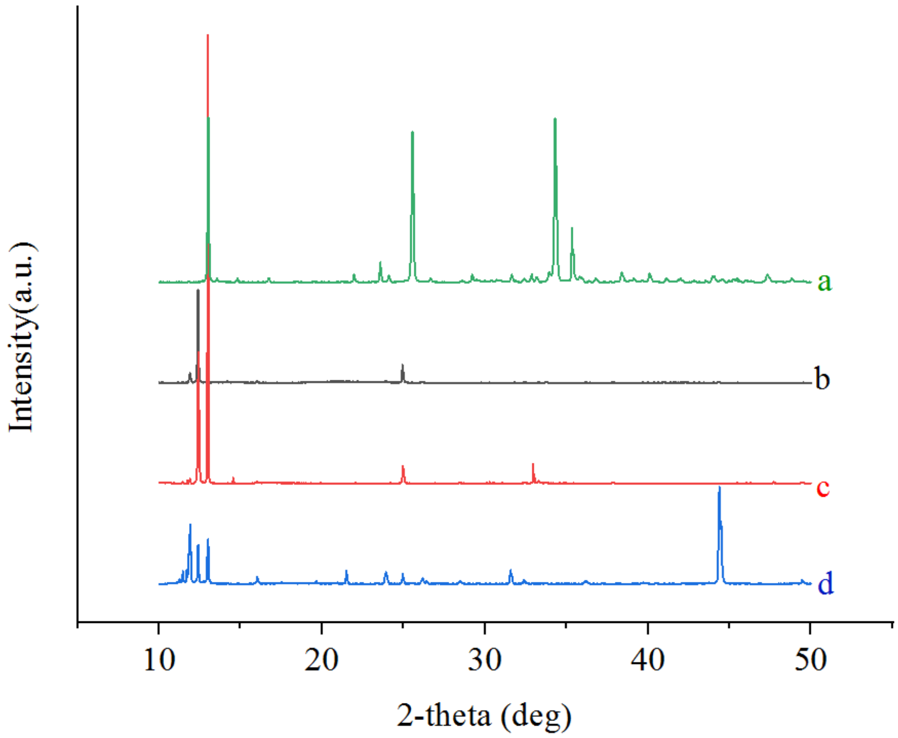


Figure 6. X-rays diffractograms obtained on:

a) sample 1: Rochelle salt flake crystal

b) sample 2: Rochelle salt film incorporated glass substrate

c) sample 3: Rochelle salt film incorporated Si substrate

d) sample 4: Rochelle salt film incorporated glass substrate

3.4 Raman measurement

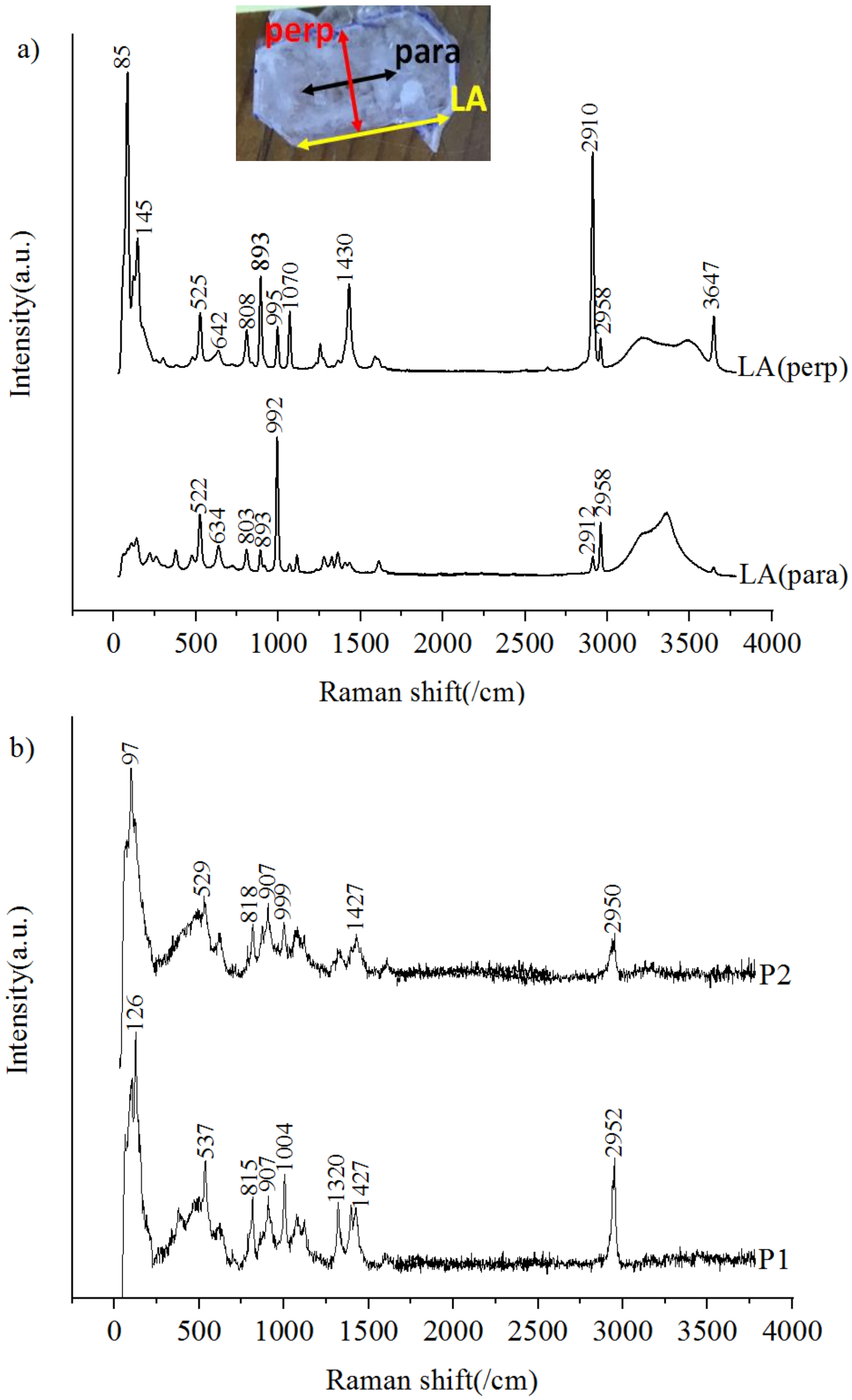
The measured Raman spectra of samples are shown in Figure 7. By comparison of Raman spectra of a single crystal of Rochelle salt in different orientations [25], the LA (perp) directed Raman spectrum of sample 1 (Figure 7a: LA means long axis direction of crystal.

Para: when the linear polarization component of the light incident on the sample is parallel to LA. Perp: when the linear polarization component of the light incident on the sample is perpendicular to LA.) is similar to the Z(YY)X direction Raman spectrum of single crystal. The LA (para) directed Raman spectrum of sample 1 is different from LA (perp) direction Raman spectrum, that is to say, in different directions there is different molecular compositions. From the above discussion can prove that the sample 1 is a single crystal structure. For sample 2, 3, 4 (Figure 7b, 7c, 7d), similar spectra can be obtained in any positions before rotating the samples 90 degrees and after rotating the samples 90 degrees (P1: the result of measurement at any position before rotating the sample 90 degrees; P2: the result of measurement at any position after rotating the sample 90 degrees). From these results, it can be concluded that all molecular compositions of Rochelle salt in each direction exist, which means that these Rochelle salt films are poly crystal structures. The peak positions of the samples Raman spectra are given in Table 3.

Table 3. The peak positions of the samples Raman spectra.

Sample 1		Sample 2		Sample 3		Sample 4	
LA (para)	LA (perp)	P1	P2	P1	P2	P1	P2
2958	2910	2952	2950	2950	2950	2954	2936
-	1430	1427	1427	1417	1422	1419	1412
992	995	1004	999	1007	1001	1007	1004
893	893	907	907	907	899	907	902
803	808	815	818	802	807	802	820
522	525	537	529	537	537	540	537
-	85	126	97	94	89	94	94

Comparing the Raman spectra of the samples with Raman spectra of a single crystal of Rochelle salt in different orientations [25], the C–H vibration gives peaks at about 2950 cm^{-1}. The broad peak at about 1412 and 1427cm^{-1} respectively, owing to the symmetric CO– vibrations. The C–C vibration gives its peaks at about 900, 990 and 1000cm^{-1}. The angle deformation of COO– mode is at around 810cm^{-1}. The torsion of COO– mode is at around 530cm^{-1} and the sharp peaks at around 90cm^{-1} are the angle deformation of CCC–, CC twist and external lattice mode. In the literatures of studies on solution-grown pure and doped sodium potassium tartrate crystals [22] and laser Raman and Infrared Spectra of Rochelle salt crystals [25], the more detailed analysis for Rochelle salt Raman spectral bands can be found.


a)
perp
para
LA
Intensity(a.u.)
85
145
525
642
808
893
995
1070
1430
2910
2958
3647
LA(perp)
992
522
634
803
893
2912
2958
LA(para)
0
500
1000
1500
2000
2500
3000
3500
4000
Raman shift(/cm)
b)
Intensity(a.u.)
97
529
818
907
999
1427
2950
P2
126
537
815
907
1004
1320
1427
2952
P1
0
500
1000
1500
2000
2500
3000
3500
4000
Raman shift(/cm)

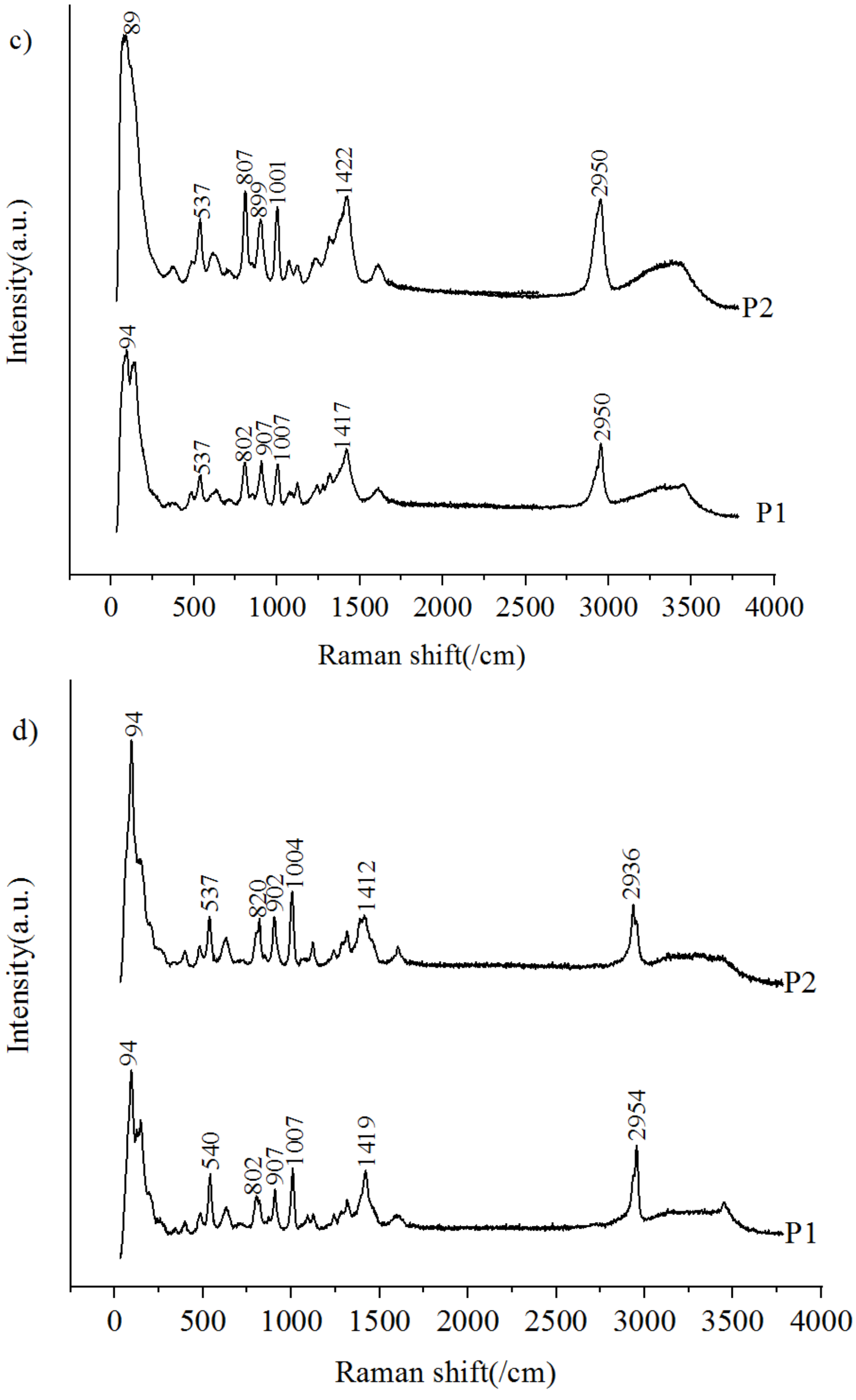


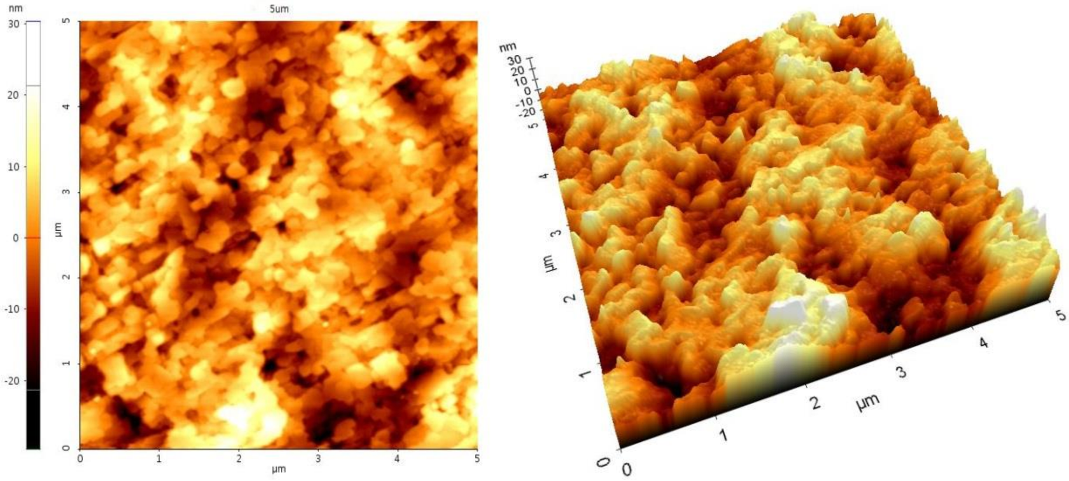
Figure 7. Raman spectroscopy patterns of: a) sample 1, b) sample 2, c) sample 3, d) sample 4.

3.5 AFM measurement

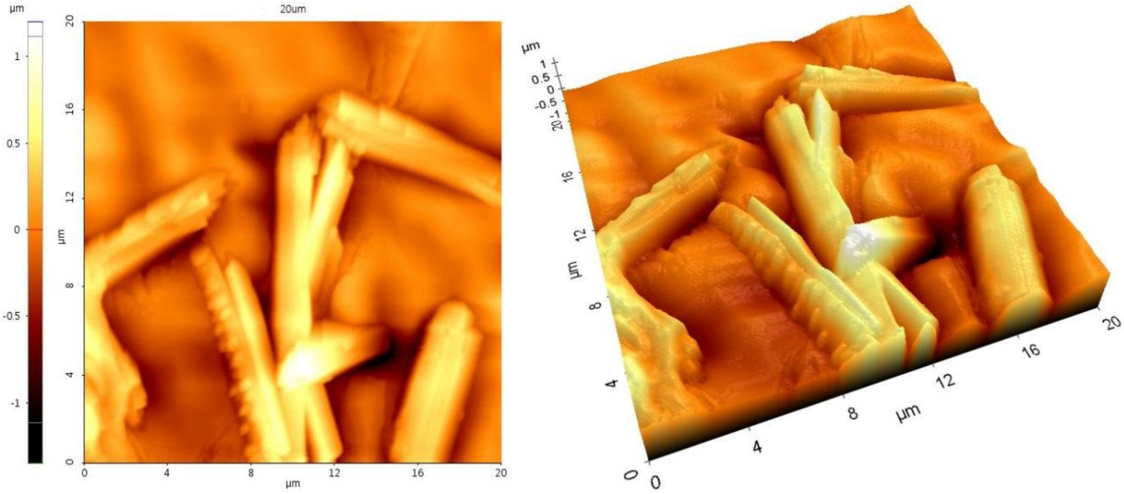
Figure 8 hows the micro scale surface morphology of samples. Neatly arranged crystal structure of simple 1 can be seen (Figure 8. For sample 3 (Figure 8), Rochelle salt crystals grown on the Si substrate, has a long rectangular shape with a narrow width and there are some single crystals with a single scattering and also have some single crystals that are combined with growth (the size of the crystal depends on the crystallization time). For sample 2 (Figure 8b) and sample4 (Figure 8d), Rochelle salt crystals grown on the glass substrate, although there are some needle-like appearance, it seems that there also has amorphous structure. The reason for the appearance of amorphous structure may be that Rochelle salt is not easily crystallized by the influence of glass or Rochelle salt crystal became amorphous for that Rochelle salt crystal was exposed to the air for some days a very thin amorphous white film appears on the surface. Thin amorphous white film appears in a day or two if the crystals are kept at too high a temperature, and suggests a slight decomposition at the surface, with perhaps some loss of water of crystallization [26].

From AFM measurement results, the crystallinity of Rochelle salt on the surface of Si substrate is better than on the surface of glass cab be found. From the result of Rochelle salt crystals grown on the Si substrate, there is a possibility of growing a single crystal film can be seen. The problem is to solve how to make a single crystal grow well in a limited space. In future work, the further studies about what materials more favorable for Rochelle salt crystallization and what structures and methods can grow crystal thin film need to be studied.

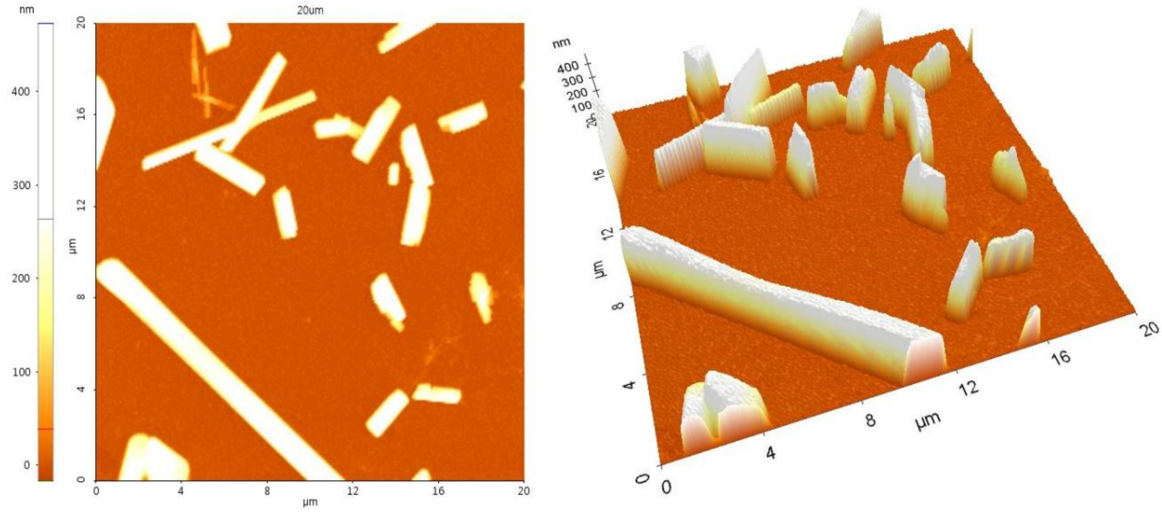
a)



b)



c)



d)

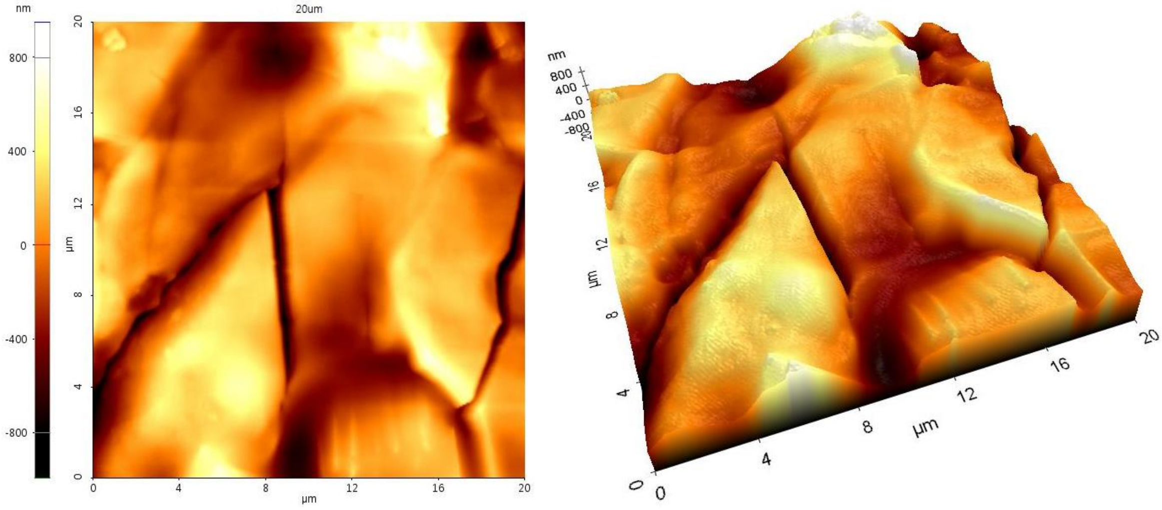


Figure 8. The AFM surface topography image of Rochelle salt crystal film samples (left is 2D, right is 3D). a) sample 1, b) sample 2, c) sample 3, d) sample 4.

3.6 Ferroelectric measurement

The measurement configuration of sample is presented in Figure 9. One side of sample was coated by Ag paste as a bottom electrode. Ag paste dots (the area is about 1mm2) were used as the electrodes of the top side of sample. The thickness of the sample is 0.9mm. The growth method of the sample is the same as the growth method of the sample 1. The measured results at different locations are shown in Figure 10. From the results, the ferroelectric properties are not presented. The reasons may be that when measuring the ferroelectric property of the Rochelle salt film grown in a limited space, which direction was measured is unclear. Rochelle salt is anisotropic and the spontaneous polarization is directed along the x crystal axis [27, 28]. Another possible reason is that the contact surface, between Ag paste and Rochelle salt film, is not good. Figure 10 shows the nonlinear curves, which may be caused by leakage current [29]. Although the ferroelectric property was not presented, the phenomenon of no closure from the hysteresis loops shows transient polarization. And the dielectric constant from this work is higher than the Rochelle salt crystal reported by B. Fugiel et al. [30].

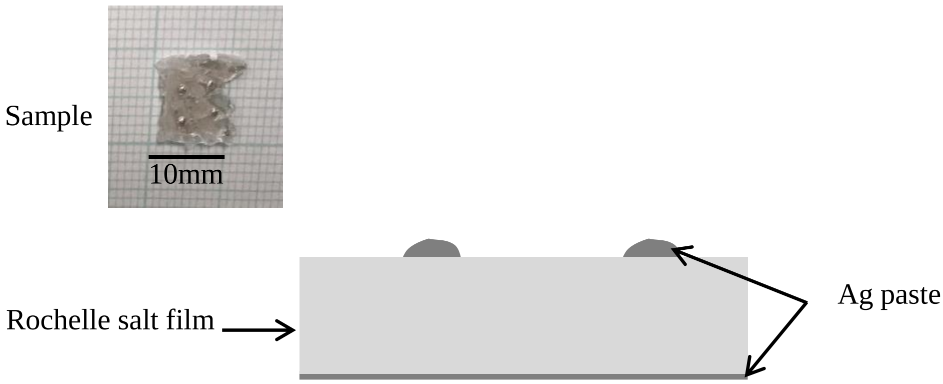


Figure 9. The schematic configuration of electrode and sample.

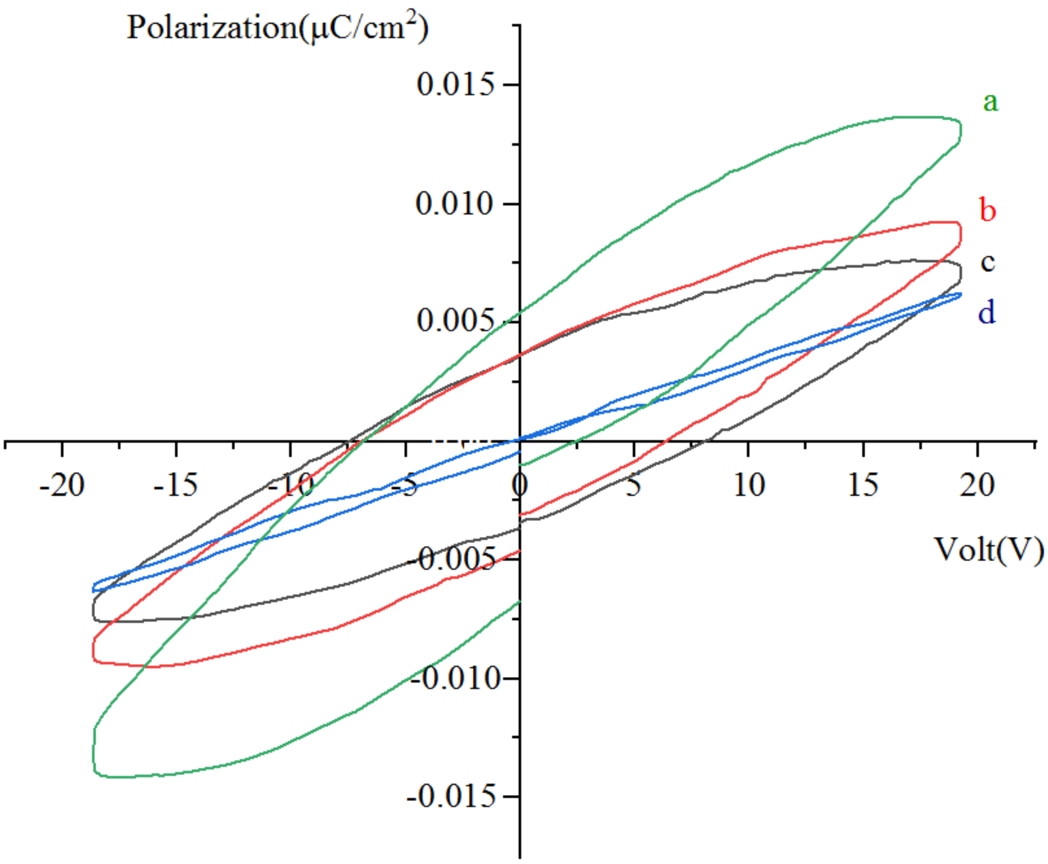


Figure 10. The measured results at different locations of the sample.

Conclusions and further work

In order to develop Rochelle salt crystal films as a medium for low operating voltage memory, fabrication of Rochelle salt thin film by evaporation solution growth method was investigated.

The obtained Rochelle salt film samples were characterized by several techniques. Through XRD and AFM measurements, it seems that these samples are crystalline. The results of Raman spectroscopy showed that all Rochelle salt samples grown in this study were crystalline. The 2mm thick sample showed single crystalline properties. However the samples below the 0.5mm thick showed polycrystalline properties. From electrical measurement, the distinct ferroelectric properties were not observed.

Although the approach in this study is favorable in principle, it takes a long time to grow. And how to control the crystal film growth into single crystalline or polycrystalline is the issue that needs to be solved. The surface morphology of crystal films grown in a limited space are better than that of crystal bulks made by cutting, but the crystalline characteristics of the surface state and the directions of the Rochelle salt also need to be studied in future work.

In order to manufacture a Rochelle salt thin films which can be applied to low operating voltage memory, we need further investigation for the microscopic arrangement and orientation of Rochelle salt crystals.

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