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Green paper-based piezoelectric material for sensors and actuators

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Abstract

In this work, the fabrication of Rochelle salt based piezoelectric structures is illustrated. Structures composed of paper and Rochelle salt are easily manufactured using simple processes. Both manufacturing and the material itself are environmental friendly. Additionally Rochelle salt is biocompatible. In the paradigm of a cleaner piezoelectric technology, the fabrication of active sensing or actuating devices is developed. Thus processing method, material and piezoelectric properties have been studied: (1) pure crystals are used as acoustic actuator, (2) properties of paper impregnated with Rochelle salt are detailed, (3) charge generation is demonstrated on the impregnated material. Actuating and sensing devices are reported in order to highlight the potential of this green piezoelectric material.

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1. Introduction

The sodium potassium tartrate tetrahydrate, $\text{NaKC}_4\text{H}_4\text{O}_6 \cdot 4\text{H}_2\text{O}$, also called Rochelle or Seignette salt is one of the ferroelectric and piezoelectric materials which include no rare element in its composition, and a good environmental compatibility. Rochelle salt is the oldest and has been for a long time the only known ferroelectric and piezoelectric material [1, 2]. Nowadays Rochelle salt is used in chemistry for different reactions especially in organic synthesis [3] and it is massively produced as a food additive (E337) also called cream of tartar [4].

Paper-based sensor [5] and piezoelectric [6] contributed recently to paper-based electronics development. In this work, the development of a green piezoelectric material has been addressed based on a method compatible with

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large area manufacturing at low-cost incorporating Rochelle salt as piezoelectric element.

2. Realization of Rochelle salt paper composite structures

In order to experiment Rochelle salt piezoelectricity, we used pure millimetric crystals as piezoelectric actuator of a paper-based cantilever (Fig. 1). Driving voltage is applied to the crystal and cantilever displacement was measured using a laser vibrometer. The dynamic first resonant mode of a paper cantilever had out-of-plane amplitude higher than 200 pm. Phase decreasing from 0 to -90° indicates that the out-of-plane displacement is the consequence of the driving signal applied to the crystal. During this experiment acoustic waves produced by the crystal could be heard by humans hears.

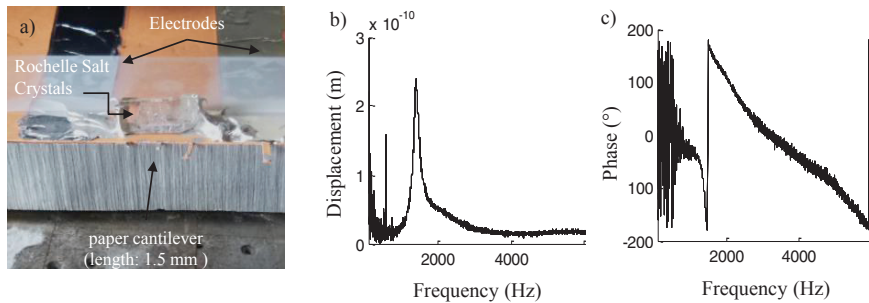


Fig. 1. (a) Array of paper cantilevers of several lengths actuated by acoustic waves produced by a Rochelle crystal fixed on the paper sheet. (b) Module and (c) Phase of the displacement of the cantilever (length 1.5 mm).

This micro-actuator concept is really efficient in terms of environmental impact, considering the system itself, its fabrication, and the material resources involved. The next logical step was to combine the salt crystal and the paper cantilever together.

In order to functionalize the paper cantilever with piezoelectric properties, an impregnation method has been developed. Once the paper is dried, Rochelle salt macro or microscopic crystal can be observed into the paper sheet depending on drying/crystallisation conditions. A homogeneous polycrystalline salt structure could be observed on Fig. 2. In this case strips of paper have been dried at ambient temperature and atmosphere into Petri dish. Then electrodes (copper tape) have been laminated on both sides of the samples.

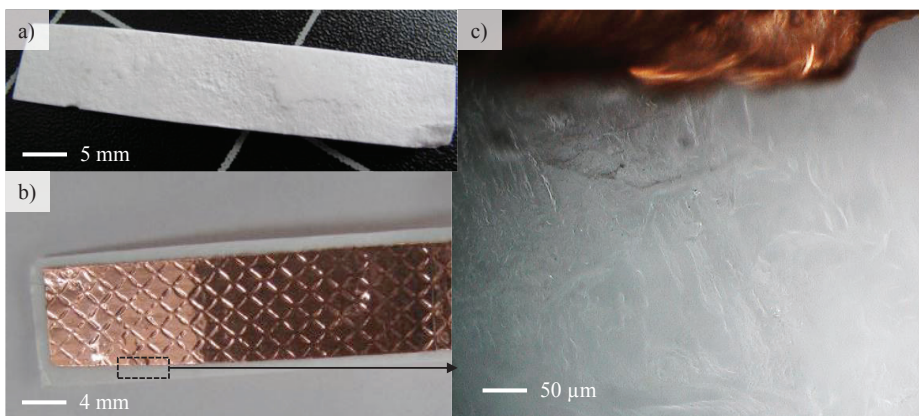


Fig. 2. (a) Paper impregnated with Rochelle salt, (b) Impregnated sample with copper tape electrodes, (c) Optical microscope image of the paper surface where the crystalline structure can be observed.

3. Characterizations

3.1. Electrical

In order to estimate the electric properties, the measurements of the capacitance and the impedance of several samples of fixed dimension have been performed. The dielectric constant and the electrical resistivity have been deduced from impedance measurements in relation with the dimensions of the samples. The results are presented in the Table 1.

Table 1. Electrical properties of paper impregnated with Rochelle salt.

Material	Dielectric constant	Resistivity ($\Omega \cdot \text{m}^{-1}$)
Short impregnation of paper	5.9	$24 \cdot 10^9$
Long impregnation of paper	8.6	$29 \cdot 10^9$
Blank paper	3.4	$56 \cdot 10^9$

The presence of Rochelle salt into paper approximatively doubles the dielectric constant of the impregnated samples. As comparison pure Rochelle salt has a dielectric constant which varies between 1100 and 9.2 depending on the measurements conditions: the dielectric constant of Rochelle salt is function of temperature and of the electric field direction with respect to crystallographic axis of the crystal [1, 7]. The dielectric constant of dry paper is typically of 2 to 4 [8] and therefore the impregnation of Rochelle salt into paper foil improved its characteristics as dielectric material.

3.2. Mechanical

Dynamic actuation of the impregnated paper cantilever was performed. A knocking experiment was carried out to verify the presence of the direct piezoelectric effect into the impregnated paper (Fig. 3). Knocking the functionalized paper cantilever produced a voltage characteristic of a damped-free oscillation. In between each impact, the paper cantilever oscillated at 143 Hz producing an electrical AC signal of maximum 100 mV (measured over the $1\text{M}\Omega$ input load of an oscilloscope). Additionally a small negative offset voltage of 15mV was observed (this offset was also observed with the raw crystal).

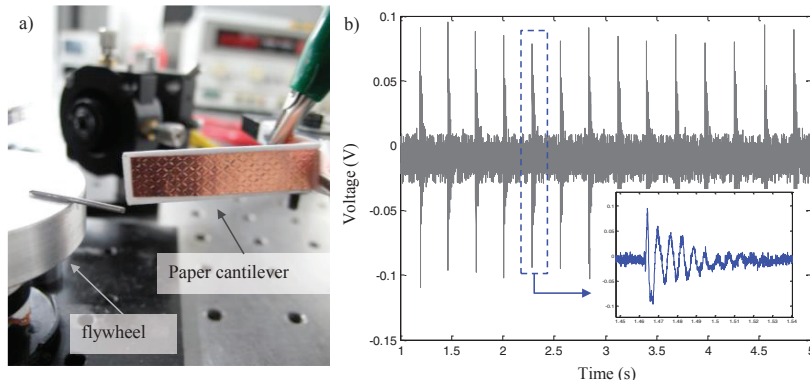


Fig. 3. (a) Impact experimental set-up consisting of the flywheel and the piezoelectric paper cantilever. (b) Electrical signals obtained from the paper cantilever recorded with an oscilloscope at each impact.

The Young modulus of the material has been deduced from the resonant behaviour of the cantilever devices. Mass density of the impregnated material has been also estimated using a precision microbalance and by measuring precisely the dimensions of the samples tested. The mechanical properties obtained are compiled in Table 2. One can notice that the impregnation of Rochelle salt into paper makes it denser and stiffer.

Table 2. Mechanical properties of paper, paper impregnated with Rochelle salt and Rochelle salt.

Material	Mass density (kg.m ⁻³)	Young Modulus (GPa)
Blank paper	551	2-4
Impregnated paper	625	7-8
Pure Rochelle salt	1770	10-18

3.3. Piezoelectric

Piezoelectric properties of the impregnated material have been measured using the Berlincourt method. Overall, temperature, electrodes adhesion, compressibility of the paper foil are several parameters that complicated the precise determination of the piezoelectric constants of the impregnated paper. Depending on the experimental conditions, the material exhibited an effective direct d_{33} coefficient measured between 3 and 25 pC.N⁻¹. To give an order of magnitude pure Rochelle salt crystal exhibits a direct piezoelectric constant that can go up to 2300 pC.N⁻¹ [9, 10] into cold environment, and between 27 and 290 pC.N⁻¹ at ambient temperature depending on the crystal orientation [11,12].

4. Conclusion and future work

In this work, the piezoelectric behavior of Rochelle salt crystal has been highlighted. Based on this, we developed a method to functionalize paper sheet with this piezoelectric material. Electric and mechanical properties of the material have been studied. Finally the direct piezoelectric effect existing into the composite material has been introduced and piezoelectricity was confirmed by using a knocking experiment. An oscillating voltage at the free-oscillation frequency of impregnated paper cantilever was measured.

This green material represents a promising technology especially well-suited for the development of large area intelligent surfaces. Smart floors, sound emitting panels or even harvesting surfaces are some of the numerous potential applications that can be envisioned at low cost. In the future, environmental sensors will be developed out of this “PiezoSalt paper”. Acoustic sensor might be the first active device that could lead to a fully biodegradable piezoelectric smart sensor.

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