



Self-writing smart labels for humidity monitoring



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ABSTRACT

Lateral anodic oxidation is a recent display technique with potential usage in self-writing smart labels and switching applications, which have the ability to operate with established RFID units. This process requires water to operate, which is harvested by a hygroscopic electrolyte layer from the ambient atmosphere. This precondition enables the realization of a humidity monitoring functionality. We develop smart labels displaying the maximum relative humidity during a period of time by the usage of halides providing different hygroscopicity in the electrolyte layer. Therefore we investigate the behavior of the humidity monitoring smart labels under test conditions with diverse relative humidity.

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1. Introduction

The self-writing mechanism caused by the process of doping front migration is a recently discovered effect and firstly introduced by Knoll [1]. These process occurs in a sandwich structure based on an activation layer composed of an intrinsically conductive polymer, typically PANI:DBSA (polyaniline doped with dodecylbenzenesulfonic acid). The optical and electrical properties are controllable by the doping state of the polymer, which is modifiable chemically or electrochemically [2–5].

In recent papers we presented a new kind of self-writing mechanism based on the self-powered anodic oxidation of a nanoscale aluminum layer laterally in one direction, carried out in an electrochemical cell, applying graphite as counter electrode [6,7]. Furthermore we introduced the facility to operate smart labels by harvesting water from the ambient atmosphere [8]. Beside the function of an opening window, we realized switching devices as an interface to electrical devices, which enable us to activate RFID units [9] and digital functions [10].

In this paper we describe a smart-label based on the lateral oxidation of nanoscale aluminum layers, for displaying irreversibly the maximum relative humidity during a period of time. The status of the label is visually detectable directly without the need of external devices.

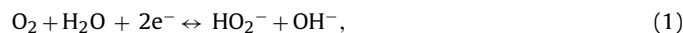
The monitoring of humidity is essential for shipping of chemicals, medical supplies and other moisture-sensitive goods. In literature is reported of humidity monitoring labels based on

RFID (radio frequency identification)-technology [11,12] or polymer films [13].

2. Process

A typical smart label based on the process of laterally anodic oxidation of nanoscale aluminum layers is shown in Fig. 1. The aluminum layer is connected to a counter electrode provides a higher standard potential than aluminum.

The cell potential is compounded of the potential of the counter electrode (−0.065 V), which is provided by the electrochemical reduction of oxygen at a graphite electrode expressed by [14]



and the potential of the pitting corrosion of aluminum (0.65–0.69 V) [15] to 0.6 V [7].

The oxidation process of the aluminum is dividable into two steps. The first step is the oxidation of elemental aluminum given by



This is followed by the hydration of the Al^{3+} -ion expressed by [16]



The aluminum layer is encapsulated partially to force the oxidation process in one dimension. This causes a moving sharp interface between the unoxidized aluminum and the residual aluminum oxide, which appears transparent.

The ionic transport is actualized by a thin electrolyte layer based on polyacrylic acid as binder to immobilize a hygroscopic halide.

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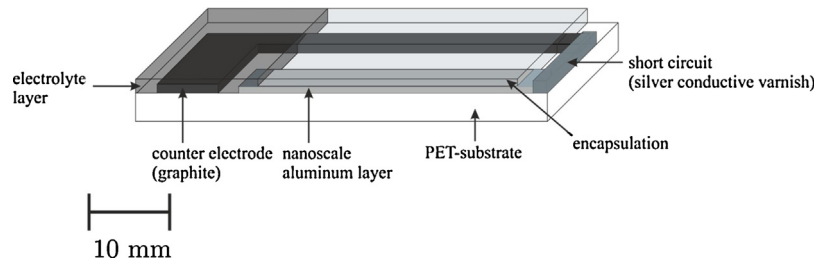


Fig. 1. Self-writing smart label with applied electrolyte layer.

The water necessary for the process is harvested by the hygroscopic halide from the ambient atmosphere. The harvesting of water from the atmosphere by a hygroscopic agent is a well known process in dehumidification application [17–21].

With the availability of water, which is a function of the ambient relative humidity, the lateral anodic oxidation takes place.

3. Fabrication

The devices were constructed from a commercially available aluminum layer (approx. 20 nm) on a polyester substrate (GBC Color Pouch, ACCO Europe). The aluminum electrode was structured in the aluminum layer by laser evaporation (Nd:YAG, $\lambda = 1064$ nm). The base material for the counter electrode consists of a mixture of acrylic resin (Wilckens) and graphite powder GSI 70 (weight ratio 3:7) and was applied by stencil printing. The devices were encapsulated by hot lamination. The typical setup of a self-writing smart label based on lateral anodic oxidation of aluminum used in this paper is depicted in Fig. 1.

The measurement of the current through the electrolyte layer at different ambient relative humidity was actualized by the measuring cell in Fig. 2 at a fixed voltage of 1.0 V DC.

For the humidity-monitoring device an arrangement of six parallel electrode units was used, which is shown in Fig. 3.

The hygroscopic electrolyte layer solution was prepared by stirring polyacrylic acid and a 1.25 M solution of the appropriate halide (weight ratio 1:19) for 60 min at room temperature. The applied

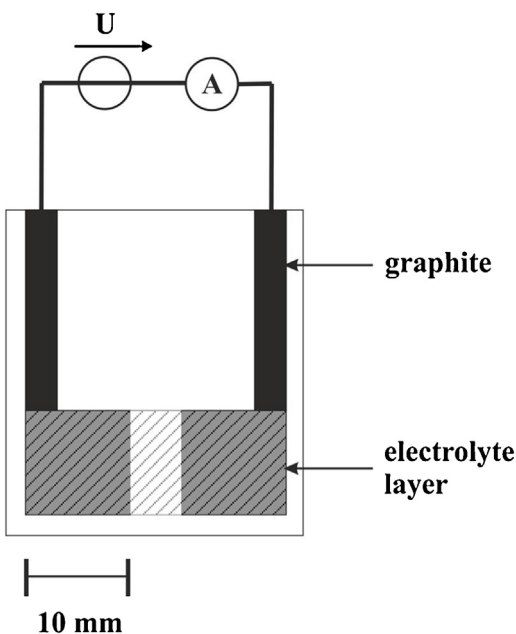


Fig. 2. Measuring cell with applied electrolyte layer based on polyacrylic acid and sodium chloride (dry thickness: 4 μ m).

halides with the resulting minimum relative humidity for operation are shown in Table 1.

Six different type of electrolyte layer (EL.1–EL.6) were applied by stencil printing with a wet thickness of 150 μ m and were dried for 24 h at relative humidity <5%.

The activation was done by short circuit at the contacts shown in the red box in Fig. 3(a).

For adjusting the relative humidity for the evaluation of the devices, saturated salt solutions were used, which provide a defined and stable equilibrium relative humidity in an enclosed container (calcium chloride: 31% [24]; potassium thiocyanate: 47% [25]; sodium bromide: 58% [23]; potassium iodide: 70% [26], sodium chloride: 75% [23]; potassium chloride: 84% [23]).

The equilibrium relative humidity controlled by hygrometer for each measurement was in agreement with literature values. The measurement of the migration lengths (l) was done automatically by a flatbed scanning device and the software package Optic Data Reader 4.1 (Christian Schoo).

4. Results and discussion

Figs. 4 and 5 depict the behavior of the migration lengths (l) of the moving interface between oxidized aluminum and aluminum dependent to the time and external relative humidity ($r.h_{ext}$).

In Fig. 4 the $l(t)$ attitude of a smart label employing sodium bromide as hygroscopic halide is shown. Therefore we observe a minimum external relative humidity of 58% for initialing the process of lateral anodic oxidation. For the case of applying sodium chloride as hygroscopic halide in the electrolyte layer we found a minimum external relative humidity for activation of 75% (Fig. 5). The observed minimum external relative humidity for operation correlates with the equilibrium relative humidity in the vapor phase above the related saturated halide solution. Therefore the minimum requirement concerning the ambient relative humidity for initializing the process of lateral anodic oxidation in a smart label based on the lateral anodic oxidation of aluminum supplied by an electrolyte layer is defined by

$$r.h_{ext} \geq r.h_{sat.sol}, \quad (4)$$

where $r.h_{ext}$ is called the external relative humidity and $r.h_{sat.sol}$ the equilibrium relative humidity of the vapor phase above a saturated solution of the related halide.

Table 1

Summary of the applied electrolyte layer type (EL.1–EL.6) with minimum relative humidity for operation dependent to the used halide.

Type	Halide	Minimum rel. humidity for operation ($r.h_{min}$)/%	Ref.
EL.1	Lithium bromide	7	[22]
EL.2	Lithium chloride	11	[23]
EL.3	Calcium chloride	31	[24]
EL.4	Sodium bromide	58	[23]
EL.5	Sodium chloride	75	[23]
EL.6	Potassium chloride	84	[23]

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