

# Control of the thermal evaporation of organic semiconductors via exact linearization

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**Abstract**—In this article, a high vacuum system for the evaporation of organic semiconductors is introduced and a mathematical model is given. Based on the exact input output linearization a deposition rate controller is designed and tested with different evaporation materials.

**Keywords**—effusion cell, organic semiconductors, deposition rate, exact linearization

## I. INTRODUCTION

THE deposition of organic semiconductors, e.g. Pentacene ( $C_{22}H_{12}$ ) and  $Alq_3$ <sup>1</sup>, becomes more and more important. Due to lower process temperatures, the manufacturing of organic electronic devices such as light emitting diodes (OLEDs) and thin film transistors (OTFTs) is less energy consuming. Additionally, these low temperatures also make it possible to use flexible substrates, e.g. plastic films, which can serve as a basis for elastic electronic devices.

The crucial part of the fabrication is the deposition of the active organic layers (films) with a layer thickness between 10 and 100 nanometres. It is mostly done by thermal evaporation in a high vacuum environment. The mobility of the charge carriers within the layers and the layer morphology strongly depend on the deposition rate, i.e. the increase in layer thickness with time during the deposition process [1], [2]. The desired deposition rates range between 0.03 and 5 Angstrom per second.

Usually, the deposition rate is controlled manually by an "expert". The result of a manually controlled deposition of Pentacene is shown in Fig. 1. The desired deposition rate is very hard to attain, the fluctuations (e.g. between 140 and 160 seconds) lead to a poor layer morphology. As a consequence of these insufficient results and the long durations of the deposition process, an automatic control of the deposition is essential.

In literature only a few control methods for the deposition of metallic materials e.g. [3] and the control of the steady state evaporation of organic materials [4] are proposed. In this work the design of a new deposition rate controller is presented. It is able to cope with transient evaporation effects. Therefore, the exact input output linearization is applied to the mathematical model [5] of the given high vacuum deposition system, described below.

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<sup>1</sup>Aluminium-tris(8-hydroxychinolin)

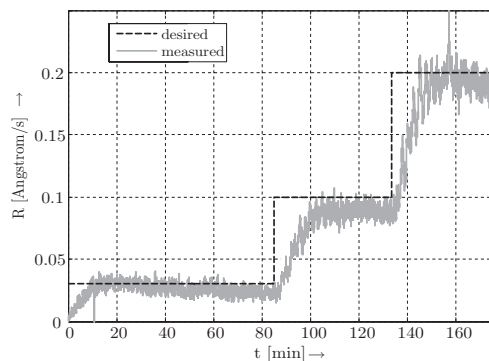


Fig. 1. Manual evaporation of Pentacene, deposition rate

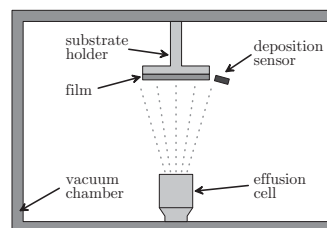


Fig. 2. Functional principle of the high vacuum system

## II. HIGH VACUUM SYSTEM

Fig. 2 shows the functional principle of the used high vacuum system. First, the vacuum chamber is evacuated by means of a rotary vane pump and a turbo molecular pump to a pressure of approximately  $10^{-6}$  mbar. The powdery evaporation material is placed in the crucible of the effusion cell (evaporation unit). By help of a resistance heating inside the effusion cell the temperature can be adjusted and the powder evaporates for sufficiently high cell temperatures. Depending on the material, the evaporation temperatures range between 100 and  $500^{\circ}C$ .

Due to the high vacuum conditions, the evaporated molecules propagate in straight lines from the effusion cell to the substrate holder. An online measurement of the deposition rate and the thickness of the deposited film is achieved by means of the deposition sensor (quartz crystal microbalance) mounted next to the substrate holder as depicted in Fig. 2.

Additionally, the transmission of the evaporation material can be stopped by closing the cell shutter (see Fig. 3). The cell is cooled by an indirect water cooling system.

As proposed in [5] the transient thermal evaporation of organic semiconductors can be described by a system of two

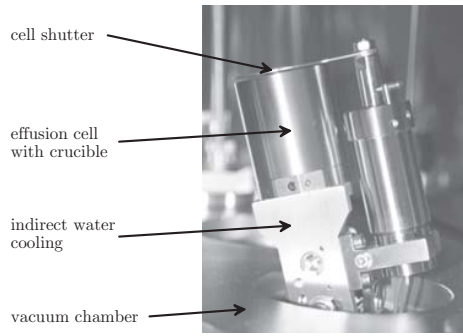


Fig. 3. Effusion cell

$\gamma_1$	$4.30 \cdot 10^{-3}$	$\delta_1$	$3.70 \cdot 10^{-2}$
$\gamma_2$	$-1.10 \cdot 10^{-10}$	$\delta_2$	$3.20 \cdot 10^{-11}$
$\gamma_3$	$-2.50 \cdot 10^{-11}$	$\delta_3$	$-2.00 \cdot 10^{-11}$
$\gamma_4$	-7.69	$\delta_4$	-1.77
$\gamma_5$	$-1.10 \cdot 10^{-1}$	$\delta_5$	$2.20 \cdot 10^{-3}$
$\gamma_6$	$1.60 \cdot 10^{-1}$	$\delta_6$	$-2.50 \cdot 10^{-3}$
		$\delta_7$	$-1.60 \cdot 10^9$
$c_1$	$2.29 \cdot 10^3$		
$c_2$	$1.40 \cdot 10^4$		

 TABLE I  
 PARAMETERS OF THE MODEL (ALQ3)

ordinary nonlinear differential equations. The state vector

$$\mathbf{x} = \begin{bmatrix} x_1 \\ x_2 \end{bmatrix} = \begin{bmatrix} T_C \\ T_M \end{bmatrix} \quad (1)$$

consists of the temperature  $T_C$  of the crucible and the evaporation temperature  $T_M$  of the organic material. The input variable  $u$  of the model

$$\begin{aligned} \frac{dx_1}{dt} &= \gamma_1 x_1^4 + \gamma_2 x_2^4 + \gamma_3 x_1 + \gamma_4 x_2 + \gamma_5 u + \gamma_6, \\ \frac{dx_2}{dt} &= \delta_1 x_1^4 + \delta_2 x_2^4 + \delta_3 x_1 + \delta_4 x_2 + \delta_5 \frac{e^{-\frac{c_2}{x_2}}}{\sqrt{x_2}} + \delta_6 u + \delta_7 \end{aligned} \quad (2)$$

equals the squared heating current of the effusion cell. The parameters  $\gamma_i$  ( $i = 1, \dots, 6$ ) and  $\delta_j$  ( $j = 1, \dots, 7$ ) are constant. The deposition rate  $R$  at the height of the substrate is the output variable of the system and can be expressed as

$$y = c_1 \frac{e^{-\frac{c_2}{x_2}}}{\sqrt{x_2}}. \quad (3)$$

The model covers the stationary evaporation in accordance with the relation of Hertz und Knudsen from kinetic theory of gases. Therefore exponential functions of the evaporation temperature  $x_2$  over the square root of the evaporation temperature appear in the mathematical descriptions (2) and (3). Furthermore the model takes into account the transient heat conductance and the reflexions of the heat radiation inside the effusion cell.

Table I lists the identified parameters of model (2), (3) for the material Alq3. These parameters yield a good match of

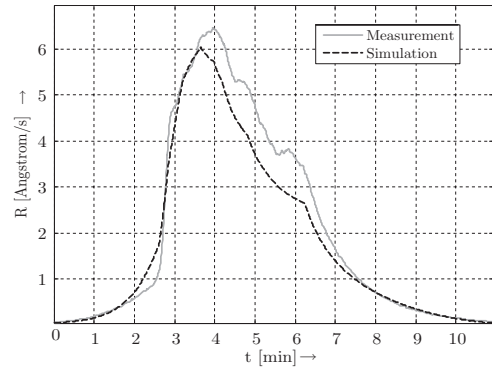


Fig. 4. Measured and simulated deposition rate (Alq3)

the measured and simulated deposition rate as shown in Fig. 4. Based on the present model a deposition controller is designed in the next section.

### III. CONTROL CONCEPT

#### A. Exact input output linearization

First, the main idea of exact input output linearization (see e.g. [7], [6]) of an  $n^{\text{th}}$ -order affine input system

$$\begin{aligned} \frac{d\mathbf{x}}{dt} &= \mathbf{a}(\mathbf{x}) + \mathbf{b}(\mathbf{x})u, \\ y &= c(\mathbf{x}) \end{aligned} \quad (4)$$

with scalar input  $u$ , scalar output  $y$  and state  $\mathbf{x}$  is outlined. The vector fields  $\mathbf{a}(\mathbf{x})$ ,  $\mathbf{b}(\mathbf{x})$  and the output function  $c(\mathbf{x})$  are assumed to be smooth.

System (4) have the relative degree  $\rho$ , if the input  $u$  appears in the  $\rho^{\text{th}}$  time-derivative of  $y$  for the first time. The use of the Lie derivative

$$L_{\mathbf{a}} c(\mathbf{x}) = \frac{\partial c}{\partial \mathbf{x}} \mathbf{a}(\mathbf{x}) \quad (5)$$

and the higher derivatives

$$L_{\mathbf{a}}^k c(\mathbf{x}) = L_{\mathbf{a}} (L_{\mathbf{a}}^{k-1} c(\mathbf{x})) , \quad L_{\mathbf{a}}^0 c(\mathbf{x}) = c(\mathbf{x}) \quad (6)$$

yields

$$\frac{d^\rho y}{dt^\rho} = L_{\mathbf{a}}^\rho c(\mathbf{x}) + L_{\mathbf{b}} (L_{\mathbf{a}}^{\rho-1} c(\mathbf{x})) u. \quad (7)$$

The relation

$$\frac{d^\rho y}{dt^\rho} \stackrel{!}{=} v \quad (8)$$

leads to a nonlinear compensation

$$u = \frac{1}{L_{\mathbf{b}} (L_{\mathbf{a}}^{\rho-1} c(\mathbf{x}))} (-L_{\mathbf{a}}^\rho c(\mathbf{x}) + v) \quad (9)$$

as shown in Fig. 5. The combination of compensation and plant results in an integrator chain of  $\rho$  integrators with input variable  $v$  and output variable  $y$ . Obviously, the state vector  $\mathbf{x}$  is required.

For the model of the high vacuum system of order  $n = 2$ , the output equation (3) solely depends on the state variable

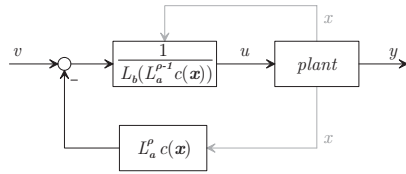


Fig. 5. Plant with nonlinear compensation

$x_2$ . The input variable  $u$  appears in the first time-derivative of  $y$ , that is

$$\frac{dy}{dt} = -\frac{c_1 e^{-\frac{c_2}{x_2}} (-2c_2 + x_2)}{2x_2^{\frac{5}{2}}} (\delta_1 x_1^4 + \delta_2 x_2^4 + \delta_3 x_1 + \delta_4 x_2 + \delta_5 \frac{e^{-\frac{c_2}{x_2}}}{\sqrt{x_2}} + \delta_6 u + \delta_7). \quad (10)$$

Therefore, the relative degree  $\rho$  equals 1. The relation

$$\frac{dy}{dt} \stackrel{!}{=} v \quad (11)$$

yields the nonlinear compensation

$$u = \frac{2x_2^{\frac{5}{2}}}{c_1 e^{-\frac{c_2}{x_2}} (-2c_2 + x_2) \delta_6} \left( -v - \delta_1 x_1^4 - \delta_2 x_2^4 - \delta_3 x_1 - \delta_4 x_2 - \delta_5 \frac{e^{-\frac{c_2}{x_2}}}{\sqrt{x_2}} - \delta_7 \right). \quad (12)$$

Due to  $\rho = 1 < n = 2$  the compensated plant has got an internal dynamics of order 1. In order to investigate this internal dynamics the zero dynamics is computed by zeroing the plant output, i.e.  $y = 0$  and  $\frac{dy}{dt} = 0$ . As a consequence the second state variable  $x_2$  equals 0 according to output equation (3). This yields

$$\delta_6 \frac{dx_1}{dt} = (\gamma_3 \delta_6 - \gamma_5 \delta_3) x_1 + (\gamma_1 \delta_6 - \gamma_5 \delta_1) x_1^4 + (\gamma_6 \delta_6 - \gamma_5 \delta_7) \quad (13)$$

for the zero dynamics. After the calculation of the equilibrium points  $x_E$  of (13) the state transformation

$$x_1 = x_E + \Delta x_1 \quad (14)$$

leads to the differential equation

$$\begin{aligned} \delta_6 \frac{d\Delta x_1}{dt} = & \gamma_1 \delta_6 x_E^4 + 4\gamma_1 \delta_6 x_E^3 \Delta x_1 + \\ & 6\gamma_1 \delta_6 x_E^2 \Delta x_1^2 + 4\gamma_1 \delta_6 x_E \Delta x_1^3 + \gamma_1 \delta_6 \Delta x_1^4 + \\ & \gamma_6 \delta_6 - \gamma_5 \delta_1 x_E^4 - 4\gamma_5 \delta_1 x_E^3 \Delta x_1 - 6\gamma_5 \delta_1 x_E^2 \Delta x_1^2 - \\ & 4\gamma_5 \delta_1 x_E \Delta x_1^3 - \gamma_5 \delta_1 \Delta x_1^4 + \gamma_3 \delta_6 x_E + \gamma_3 \delta_6 \Delta x_1 - \\ & \gamma_5 \delta_3 x_E - \gamma_5 \delta_3 \Delta x_1 - \gamma_5 \delta_7 \end{aligned} \quad (15)$$

for the displacement  $\Delta x_1$ . A quadratic Lyapunov function

$$V(\Delta x_1) = \frac{1}{2} \Delta x_1^2 \quad (16)$$

is used to determine the asymptotic stability of  $x_E$ . Its time derivative  $\frac{dV}{dt}$  is locally negative as it is shown in Fig. 6. As a consequence the zero dynamics is asymptotically stable and the compensated plant is minimum-phase. Therefore, the exact linearization technique can be applied in the present case.

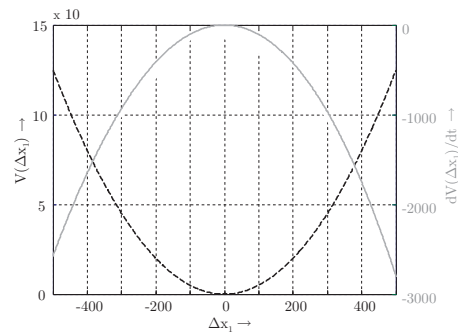


Fig. 6. Lyapunov function for the zero-dynamic

	Pentacene	Alq3
$K_P$	$5 \cdot 10^{-2}$	$3 \cdot 10^{-1}$
$K_I$	$2 \cdot 10^{-3}$	$1 \cdot 10^{-2}$

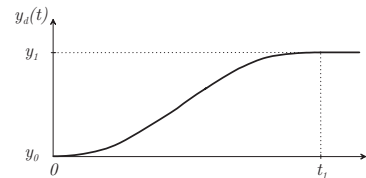
 TABLE II  
PARAMETERS OF THE LINEAR CONTROLLER


Fig. 7. Desired trajectory for the deposition rate

### B. Controller design

To control the compensated plant, a PI-controller

$$C(s) = K_P + \frac{K_I}{s} \quad (17)$$

is used. Its parameters  $K_P$  and  $K_I$  are constant and were found by simulation. Table II shows the chosen control parameters for the materials Pentacene and Alq3.

The implementation of the control algorithm in the high vacuum system is done in the discrete time form [8]

$$u_i = u_{i-1} + K_P e_i + (K_I T_S - K_P) e_{i-1}, \quad (18)$$

with a sampling period  $T_S$ . Here,  $u_i$  symbolises the actuating signal and  $e_i$  the control error at the time instants  $t = iT_S$  ( $i = 0, 1, 2, \dots$ ). In order to prevent the windup-effect, the integral action is turned off in case of a saturated actuating signal.

### C. Trajectory planning

A trajectory for a transition between a initial point  $y(t=0) = y_0$  at time  $t = 0$  and an final point  $y(t=t_1) = y_1$  at time  $t_1$  should be planned as shown in Fig 7. Therefore, the polynomial [9]

$$y_d(t) = y_0 + (y_1 - y_0) \sum_{i=n+1}^{2n+1} p_i \left( \frac{t}{t_1} \right)^i \quad (19)$$

is chosen. If the first  $n$  derivatives of  $y_d$  should vanish at  $t = 0$  and  $t = t_1$ , the parameters  $p_i$  can be evaluated uniquely.

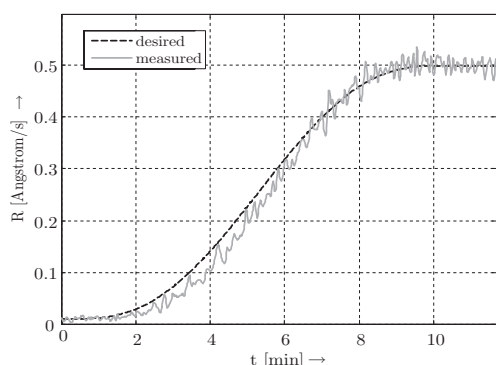


Fig. 8. Control of the deposition rate (Pentacene)

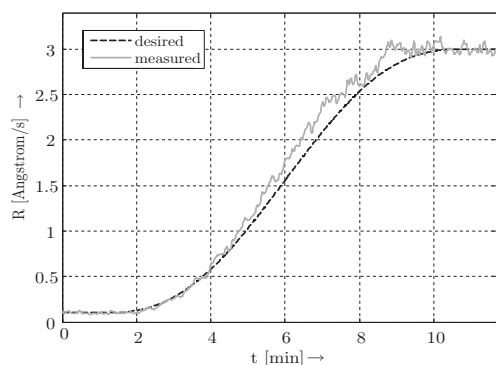


Fig. 9. Control of the deposition rate (Alq3)

#### IV. RESULTS

As depicted in Fig. 8 the proposed control approach based on the exact input output linearization leads to outstanding results for the evaporation of Pentacene. In contrast to the deposition by hand and the approach in [4] also transients can be tracked in a very satisfactory manner.

Even for Alq3 the proposed method yields highly satisfying results as shown in Fig. 9. Note that Alq3 is more difficult to evaporate as it tends to impulsive expulsion of the material during a heating process.

#### V. CONCLUSION

A high vacuum system for evaporating organic semiconductors is introduced and a mathematical model consisting of two ordinary nonlinear differential equations is given to describe the systems behaviour. Based on this model a nonlinear compensation is designed by means of exact linearization technique.

The nonlinear compensation and a PI-Controller are used to control the deposition rate during the evaporation. It is shown that the method is well-suited to solve this task.

As the main drawback of this approach, the state variables have to be measurable. Other nonlinear control methods e.g. a flatness based approach can be used to overcome this difficulty.

#### ACKNOWLEDGMENT

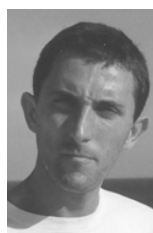
This work was funded by the Austrian Research Promotion Agency (FFG, no. 828647). Special thanks to the members of the Institute for Surface Technologies and Photonics of Joanneum Research Forschungsgesellschaft mbH, especially Alexander Fian and Georg Jakopic, for their help and contributions to this work.

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