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## Modeling resistance increase in a composite ink under cyclic loading

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# Flexible and Printed Electronics



## PAPER

# Modeling resistance increase in a composite ink under cyclic loading

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Q Li , E Chung , A Antoniou\* and O Pierron\*

G.W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, GA 30318, United States of America

\* Authors to whom any correspondence should be addressed.

E-mail: [antonia.antoniou@me.gatech.edu](mailto:antonia.antoniou@me.gatech.edu) and [olivier.pierron@me.gatech.edu](mailto:olivier.pierron@me.gatech.edu)

**Keywords:** conductive inks, cyclic stretching, failure lifetime, modeling

Supplementary material for this article is available [online](#)

## Abstract

The electrical performance of stretchable electronic inks degrades as they undergo cyclic deformation during use, posing a major challenge to their reliability. The experimental characterization of ink fatigue behavior can be a time-consuming process, and models allowing accurate resistance evolution and life estimates are needed. Here, a model is proposed for determining the electrical resistance evolution during cyclic loading of a screen-printed composite conductive ink. The model relies on two input specimen-characteristic curves, assumes a constant rate of normalized resistance increase for a given strain amplitude, and incorporates the effects of both mean strain and strain amplitude. The model predicts the normalized resistance evolution of a cyclic test with reasonable accuracy. The mean strain effects are secondary compared to strain amplitude, except for large strain amplitudes ( $>10\%$ ) and mean strains ( $>30\%$ ). A trace width effect is found for the fatigue behavior of 1 mm vs 2 mm wide specimens. The input specimen-characteristic curves are trace-width dependent, and the model predicts a decrease in  $N_f$  by a factor of up to 2 for the narrower trace width, in agreement with the experimental results. Two different methods are investigated to generate the rate of normalized resistance increase curves: uninterrupted fatigue tests (requiring  $\sim 6-7$  cyclic tests), and a single interrupted cyclic test (requiring only one specimen tested at progressively higher strain amplitude values). The results suggest that the initial decrease in normalized resistance rate only occurs for specimens with no prior loading. The minimum-rate curve is therefore recommended for more accurate fatigue estimates.

## 1. Introduction

Flexible hybrid electronics devices have attracted a lot of research interest in recent years due to their many applications, including wearable health-care monitoring [1–4], energy storage [5, 6], flexible displays [7–9], and implantable bioelectronics [10, 11]. These devices integrate electronic components with compliant electric circuits based on interconnects that can maintain electrical conductivity under repeated deformation, such as repeated elongation. In wearable device applications, repeated elongation (stretching) of up to 30% strain is typically expected [12]. Various conductive interconnect materials have been studied for their electrical performance under

repeated deformation, including thin metal films [13–17], metal nanoparticle inks [18–21], and metal-polymer composite inks [22–28]. One class of metal-polymer composite ink (conductive ink) consists of micron-sized metal flakes embedded in a compliant polymer binder material. The conductive ink is then deposited onto a compliant polymer substrate, typically by screen or gravure printing. The current work used the PE 874 conductive ink provided by DuPont, a stretchable ink with silver flakes embedded in a polyurethane binder and screen-printed on a thermoplastic polyurethane substrate.

As the PE 874 conductive ink is stretched, strain localization occurs in the ink by surface cracking starting from very low strains ( $\sim 1\%$ ) [28, 29]. In both

the monotonic and cyclic cases, the evolving crack pattern is linked to a resistance increase. From *in-situ* confocal microscope (CM) and scanning electron microscope (SEM) experiments, the surface cracks lengthen as well as deepen and widen with monotonic straining corresponding to a steady increase in electrical resistance up to applied strains of  $\sim 150\%$  [28]. When the ink is cyclically strained between two strain values, the maximum electrical resistance during a cycle also increases with cycling. The *in-situ* CM and SEM cyclic experiments showed that although the extent of the crack pattern does not change with cyclic straining, the cracks that exist at the maximum strain during the first cycle deepen and widen with further cycling [28].

Characterization and modeling of the electrical fatigue behavior of these inks is crucial to design functional stretchable electronics devices with proper reliability. One way to characterize fatigue of a conductive ink specimen is by defining a fatigue life  $N_f$  as the number of cycles until a specified normalized resistance  $R/R_0$  (with respect to the initial resistance  $R_0$ ) is reached, and measuring  $N_f$  for different strain amplitudes. Previous work on the PE 874 ink employed critical normalized values of 500 [22] or 100 [28] to define  $N_f$ . In each case, a strain amplitude  $\varepsilon_a$ –fatigue life  $N_f$  curve is obtained [22, 28]. The advantage of these curves is that they provide estimates  $N_f$  for any given  $\varepsilon_a$ , using empirical fits relating  $N_f$  to  $\varepsilon_a$  from discrete experimental datasets. However, there are limitations to this approach. First, the  $\varepsilon_a$ – $N_f$  curve should be obtained for a fixed value of mean strain ( $\varepsilon_m$ ). Even though previous work [28] showed that  $\varepsilon_a$  has a much more significant effect on  $N_f$  than  $\varepsilon_m$ , the effect of  $\varepsilon_m$  cannot always be neglected (especially in the case of large  $\varepsilon_m$  values). Proper characterization would require obtaining several  $\varepsilon_a$ – $N_f$  curves for different  $\varepsilon_m$  values. More importantly, the empirical fits obtained from  $\varepsilon_a$ – $N_f$  curves do not provide any indication of the increase in  $R/R_0$  during cycling. If a different definition of  $N_f$  is to be used (say for  $R/R_0 = 50$ , or 10), a new set of curves needs to be generated. As such, modeling the resistance increase during cycling for any combination of  $\varepsilon_a$  and  $\varepsilon_m$  would be much more valuable, which this paper addresses.

Li *et al* [28] showed that the rate of normalized resistance change with cycling,  $d(R/R_0)/dN$ , or  $(R/R_0)'$  for shorthand, is dictated by  $\varepsilon_a$ . More specifically,  $(R/R_0)'$  can be characterized by two values (initial maximum, and minimum values), which were shown to have a strong correlation with  $\varepsilon_a$  (whereas the effect of  $\varepsilon_m$  was found to be negligible). The current work presents a model for estimating the  $R/R_0$  evolution during cyclic loading. Figure 1 shows the overall modeling approach in a schematic. The model is based on two input specimen-characteristic curves: either of the  $(R/R_0)'$ – $\varepsilon_a$  curves from cyclic uniaxial test(s) and the  $R/R_0$ – $\varepsilon$  curve from a monotonic uniaxial test.

Importantly, the model accounts for both  $\varepsilon_a$  and  $\varepsilon_m$ . From these inputs, the  $R/R_0$  evolution over cycles can be predicted using the model. During each cycle,  $R/R_0$  shifts between a maximum and minimum corresponding to the maximum and minimum strain values respectively, and the maximum  $R/R_0$  measured during a cycle is referred to as  $R_{\max}/R_0$  (see examples in figures 2(d) and 3(a)). The predicted  $R_{\max}/R_0$  using the model is referred to as  $\hat{R}_{\max}/R_0$ . Modeling the  $\hat{R}_{\max}/R_0$  evolution over cycles also means that the fatigue life can be predicted—by calculating the number of cycles to reach  $\hat{R}_{\max}/R_0 = 100$ . The predicted fatigue life is referred to as  $\hat{N}_f$ . Based on the input curves, the  $\varepsilon_a$ – $\hat{N}_f$  curves can be generated for different values of  $\varepsilon_m$ . Therefore, the outputs of the model are the  $\hat{R}_{\max}/R_0$  evolution over cycles and  $\hat{N}_f$ .

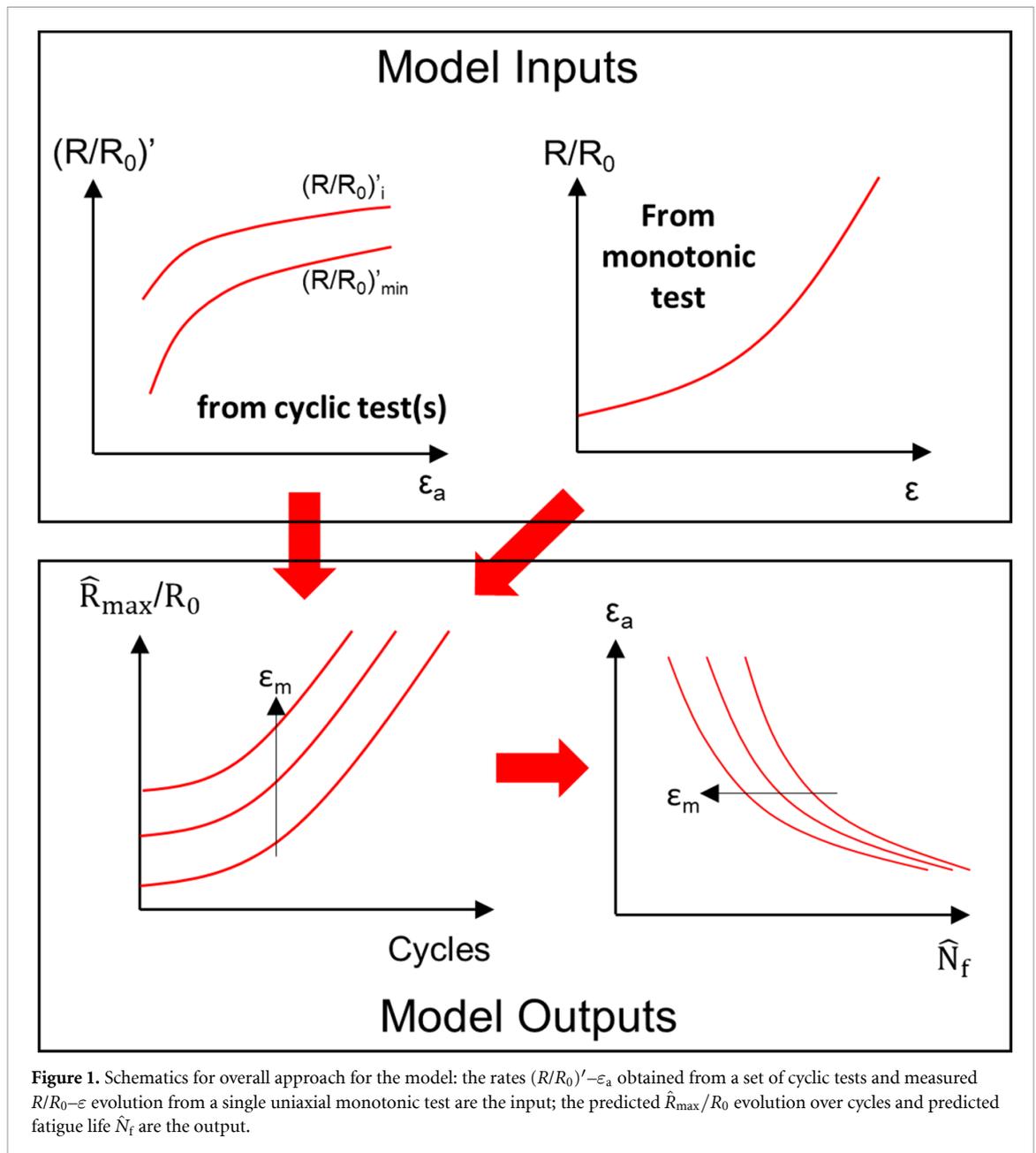
The model is first applied to the 2 mm-wide-specimens fatigue data from Li *et al* [28]. Additional fatigue data are also presented and modeled for a narrower, 1 mm-ink trace width, in order to further assess the effect of trace width on the fatigue behavior [22, 28]. An experimental procedure for obtaining the  $(R/R_0)'$ – $\varepsilon_a$  curve using only one specimen (by performing a series of interrupted cyclic tests at progressively higher  $\varepsilon_a$ ) is also investigated and compared to the procedure where a different specimen is used for each  $\varepsilon_a$ .

## 2. Experimental and modeling methods

### 2.1. Experimental procedures

#### 2.1.1. Fabrication of ink specimens

The PE 874 conductive ink formulated by DuPont is composed of silver flakes embedded in a polyurethane binder material. The average volume fraction of silver flakes, whose sizes range from several  $\mu\text{m}$  to 100s of  $\mu\text{m}$ , is about 55% [30]. The PE 874 ink test specimens used in this work consist of two layers of the PE 874 ink screen printed onto a thermoplastic polyurethane (TPU) substrate layer in two separate passes. The TPU used for the substrate is the TE-11C from DuPont. The screen-printing process was performed at the DuPont Applications Laboratory with proprietary processes that have been optimized for the ink and substrate. For the first ink layer, a mesh size of 325 threads crossing per inch<sup>2</sup> with wire diameter of 0.9 mil was used. For the second ink layer, a mesh size of 280 threads crossing per inch<sup>2</sup> with wire diameter of 1.2 mil was used. For all cases, the mesh angle was 30°. There was a 15 min drying time after the printing of each ink layer, at 125 °C for the first ink layer and 130 °C for the second layer. The ink is printed in U-shaped, double trace lines with 2 mm or 1 mm trace width (figure 2(a)). The four pads in the print pattern were designed for the four-point electrical resistance probes. The average total thickness of the two ink layers measured by DuPont was 20  $\mu\text{m}$ . The average thickness of the TPU substrate is 127  $\mu\text{m}$ .



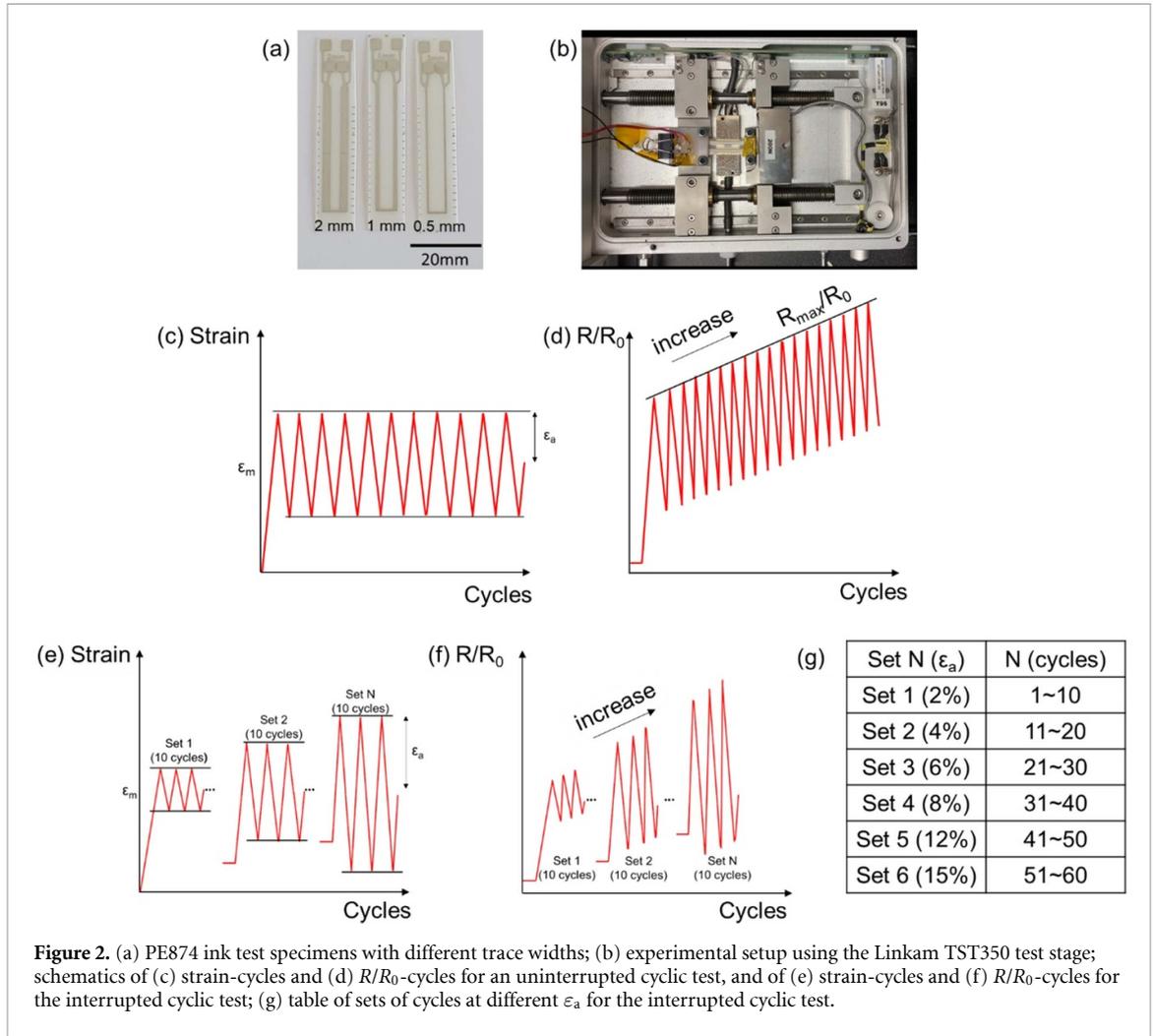
**Figure 1.** Schematics for overall approach for the model: the rates  $(R/R_0)'$ – $\varepsilon_a$  obtained from a set of cyclic tests and measured  $R/R_0$ – $\varepsilon$  evolution from a single uniaxial monotonic test are the input; the predicted  $\hat{R}_{\max}/R_0$  evolution over cycles and predicted fatigue life  $\hat{N}_f$  are the output.

### 2.1.2. Cyclic test procedures

The cyclic uniaxial stretching tests were performed with synchronous electrical resistance measurements on the Linkam Scientific TST350 Microtensile Test Stage (figure 2(b)) at a strain rate of 2% per second. The electrical resistance is measured using the Agilent 34401A digital multimeter. The cyclic stretching tests were performed by first stretching the test specimen to the maximum strain  $\varepsilon_m + \varepsilon_a$  (mean strain plus strain amplitude), and then cycling between the maximum strain  $\varepsilon_m + \varepsilon_a$  and minimum strain  $\varepsilon_m - \varepsilon_a$ . In addition, a series of interrupted cyclic tests were performed, in order to obtain relevant rate parameters for a range of  $\varepsilon_a$ 's using only one specimen. For the interrupted tests firstly, a cyclic test was performed at a low  $\varepsilon_a$  (2% in the current work) for ten cycles, then the test specimen was unloaded. Next

another test was performed at a higher  $\varepsilon_a$  on the same specimen for only ten cycles, then the specimen was again unloaded. This process is repeated for progressively higher  $\varepsilon_a$  up to 18%. Figures 2(c)–(f) show schematics of the strain and  $R/R_0$  evolution over cycles for an uninterrupted (figures 2(c)–(d)) versus interrupted (figures 2(e)–(f)) test. A table of the sets of cycles used for the interrupted test is shown in figure 2(g).

The resistance is reported as the normalized value  $R/R_0$ , where  $R$  is the current resistance and  $R_0$  is the initial resistance before testing. Due to the distance  $d_{\text{clamp}}$  between the specimen clamps (about 30 mm) being shorter than half the overall length  $l_{\text{print}}$  of the double trace line (38 mm), the initial resistance  $R_0$  needed to be adjusted for the unstrained portion of the specimen:



$$R_0 = R_{\text{full specimen}, \epsilon=0} \times \left( \frac{d_{\text{clamp}}}{l_{\text{print}}/2} \right) \quad (1)$$

The resistance  $R$  is the sum of the initial resistance  $R_0$  and the measured change in resistance  $\Delta R$ , which is entirely attributed to the strained portion of the specimen:

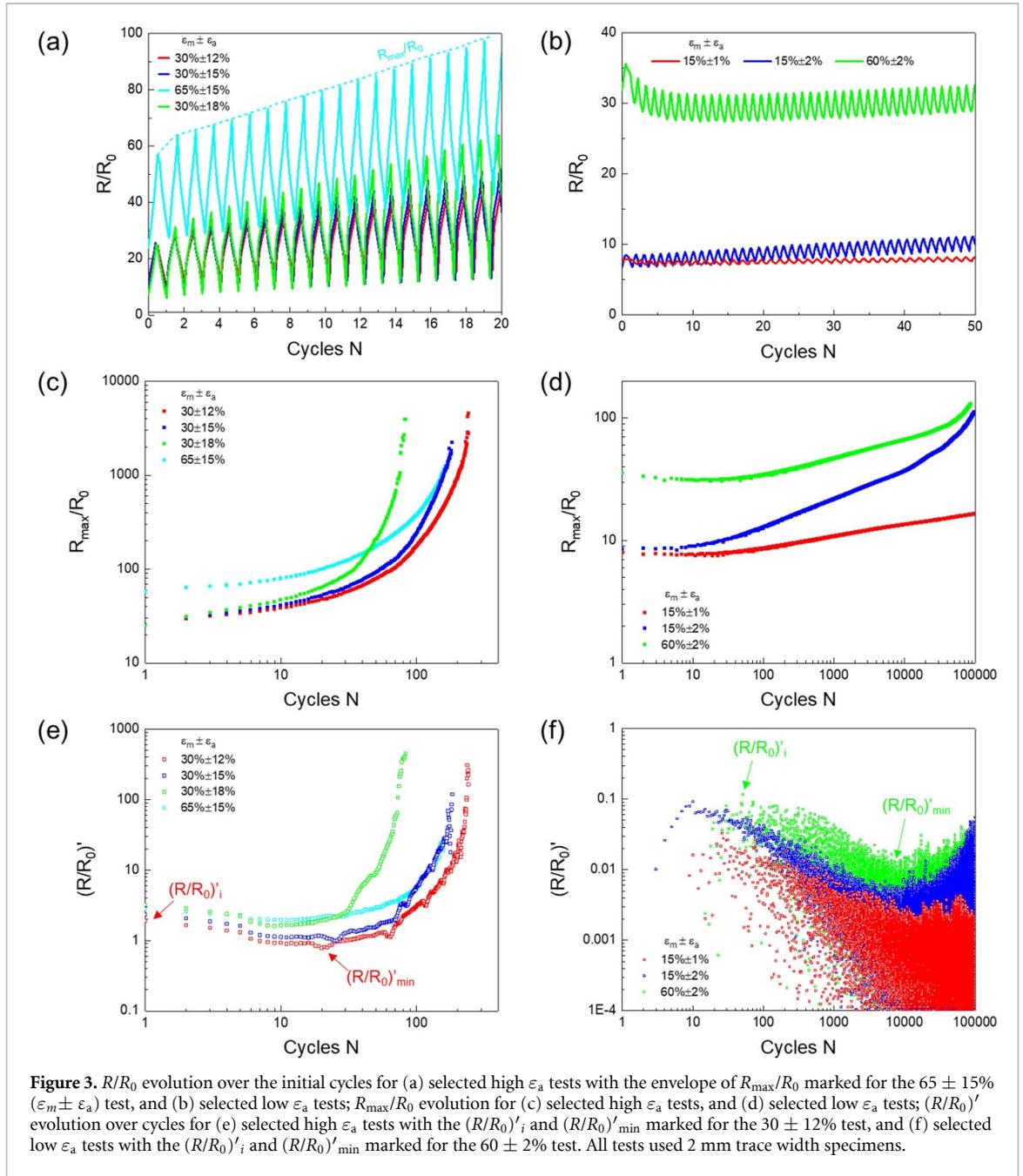
$$\Delta R = R_{\text{full specimen}, \epsilon} - R_{\text{full specimen}, \epsilon=0} \quad (2)$$

$$R = R_0 + \Delta R. \quad (3)$$

### 2.1.3. Determination of rate of normalized resistance increase

The rate of resistance change with cycling,  $(R/R_0)'$ , at any cycle  $N$  was calculated by fitting a linear regression function to the measured  $R_{\text{max}}/R_0$  evolution over cycling (see examples in figures 3(a)–(d)) between  $N - 4$  and  $N + 4$ , excluding any null points at the beginning or end of the set of cycles. The choice of using the cycles  $N - 4$  to  $N + 4$  for the linear regression fitting was made by trial to achieve the generally smooth evolution of  $(R/R_0)'$  over the cycles. The initial rate  $(R/R_0)'_i$  and minimum rate  $(R/R_0)'_{\text{min}}$  were identified in [28] as relevant rate parameters for characterizing resistance evolution with cycling.

The evolution of  $(R/R_0)'$  with cycling for 2 mm-wide-specimens is summarized in figure 3. For  $\epsilon_a \geq 3\%$ , the normalized resistance always increases with cycling ( $(R/R_0)' > 0$ ); moreover,  $(R/R_0)'$  initially decreases with cycling until reaching a minimum value, after which  $(R/R_0)'$  increases significantly (see figure 3(e)). In that large  $(R/R_0)'$  stage (where  $R/R_0$  increases by more than 5 times every cycle), the ink is not considered functional anymore. For  $\epsilon_a = 1$  or 2%, the normalized resistance tends to first decrease within the few cycles ( $(R/R_0)' < 0$ ), after which it slowly increases (see figure 3(f)). The initial decrease in  $R/R_0$  is likely associated with relaxation of the substrate (as shown in [22] when holding a specimen at constant strain), and the fact that the fatigue damage is not significant enough to result in a large  $R/R_0$  increase to counteract the small relaxation effects. As a result, the  $(R/R_0)'$  is maximum at cycle  $\sim 10$ –50, then decreases with cycling until reaching a steady-state minimum value. The initial rate  $(R/R_0)'_i$  is defined by the initial local maximum in the  $(R/R_0)'$  evolution. For  $\epsilon_a \geq 3\%$ ,  $(R/R_0)'_i$  was found to occur at cycle 1; for  $\epsilon_a = 1\%$  or 2%,  $(R/R_0)'_i$  occurs at a later cycle but within the first 50 cycles for the aforementioned reason (see figure 3(f)). The minimum rate  $(R/R_0)'_{\text{min}}$



is defined by the absolute minimum  $(R/R_0)'$  for cases with  $\varepsilon_a \geq 10\%$  or the minimum of the upper bound of the  $(R/R_0)'$  data for cases with  $\varepsilon_a < 10\%$ , given the large spread in the  $(R/R_0)'$  data for these cases (see figure 3(f)).  $(R/R_0)'_{\min}$  occurred at 10's of cycles for the high  $\varepsilon_a$  tests and 1000's of cycles for the low  $\varepsilon_a$  tests. An alternative method for obtaining the rate parameters was implemented with the interrupted test described in section 2.1.2. For that method, the minimum  $(R/R_0)'$  during a set of ten cycles (at a given  $\varepsilon_a$ ) was used as the  $(R/R_0)'_{\min}$  parameter for that  $\varepsilon_a$ .

## 2.2. Model of normalized resistance evolution under cyclic loading

A simple model is derived to predict resistance increase as a function of the number of applied cycles

$N$ . For the first cycle ( $N = 1$ ),  $\hat{R}_{\max}/R_0$  is obtained at the maximum strain value  $\varepsilon_m + \varepsilon_a$  during the cycle, which can be obtained from the monotonic data. The additional increase in  $\hat{R}_{\max}/R_0$  with cycling is assumed to be linear with the number of cycles (constant  $(R/R_0)'$ ). Using the maximum initial rate  $(R/R_0)'_i$  provides a conservative model, as follows:

$$\frac{\hat{R}_{\max}}{R_0} = \left( \frac{R_{\max}}{R_0} \right)_{N=1} + (N-1) \left( \frac{R}{R_0} \right)'_i. \quad (4)$$

A fatigue live  $\hat{N}_f$  can also be predicted. If failure is determined by  $\hat{R}_{\max}/R_0 = 100$ ,  $\hat{N}_f$  is:

$$\hat{N}_f = \frac{100 - \left( \frac{R_{\max}}{R_0} \right)_{N=1}}{\left( \frac{R}{R_0} \right)'_i} + 1. \quad (5)$$

The  $\hat{R}_{\max}/R_0$  of the first cycle as a function of  $\varepsilon_m$  and  $\varepsilon_a$  is obtained from the polynomial fit of the  $R/R_0-\varepsilon$  data from a single monotonic test. The  $(R/R_0)'_i$  as a function of  $\varepsilon_a$  is obtained from the power law function fit of the  $(R/R_0)'_i-\varepsilon_a$  data from a set of cyclic tests. The model for  $\hat{R}_{\max}/R_0$  and  $\hat{N}_f$  can be similarly formulated using  $(R/R_0)'_{\min}$ :

$$\frac{\hat{R}_{\max}}{R_0} = \left(\frac{R_{\max}}{R_0}\right)_{N=1} + (N-1) \left(\frac{R}{R_0}\right)'_{\min} \quad (6)$$

$$\hat{N}_f = \frac{100 - \left(\frac{R_{\max}}{R_0}\right)_{N=1}}{\left(\frac{R}{R_0}\right)'_{\min}} + 1. \quad (7)$$

Although the current model focuses on  $\hat{R}_{\max}/R_0$  during the cycle at the maximum strain, analogous models can be derived for strain values during the cycle other than the maximum strain. For any strain value during the cycle,  $(R/R_0)_{N=1}$  can be obtained at the strain value from a monotonic test and  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$  at the strain value can be obtained from cyclic tests in an analogous manner to those defined for  $R_{\max}/R_0$ .

### 3. Results and discussion

#### 3.1. Modeling results for 2 mm-wide specimens: $R/R_0$ evolution and $N_f$ estimates

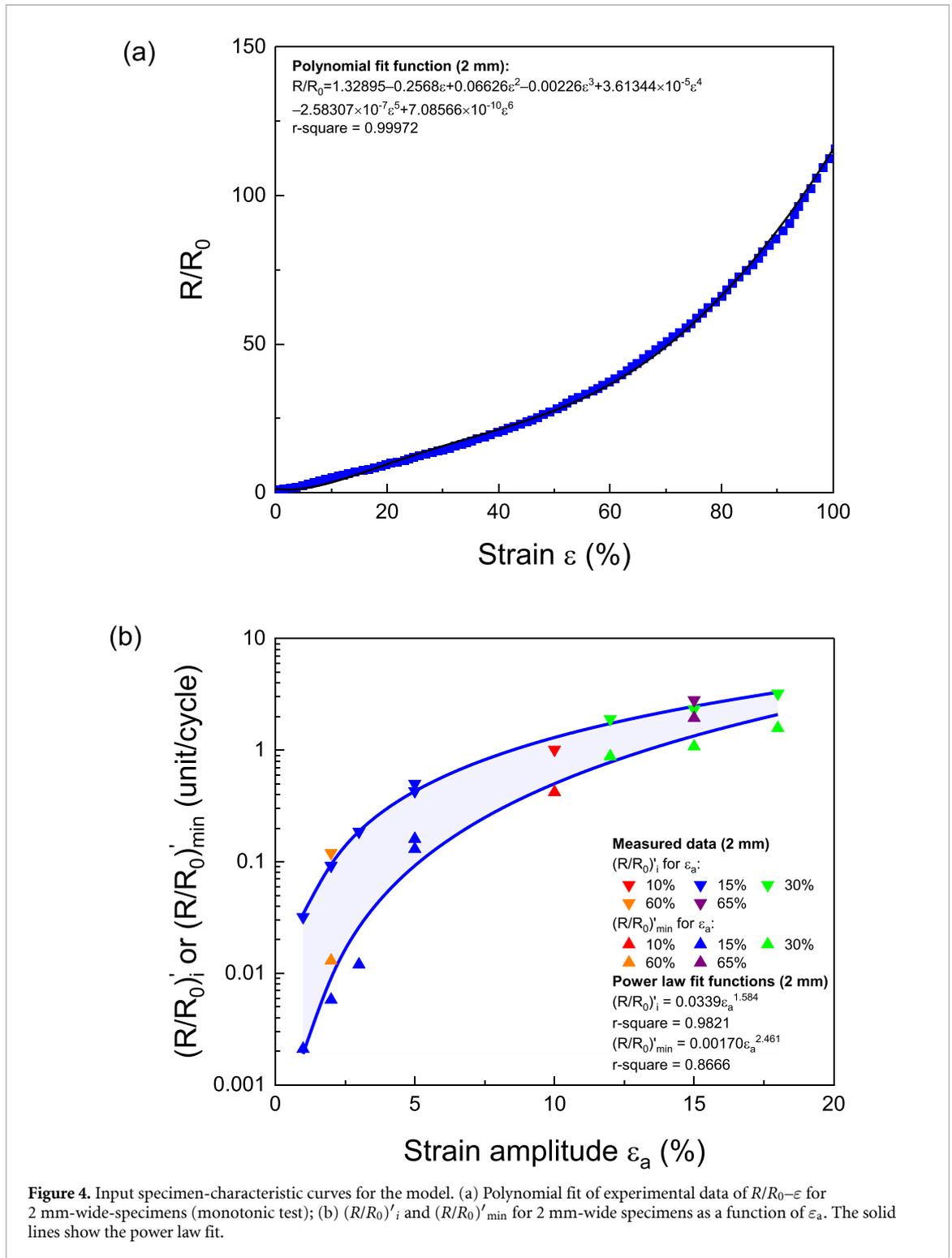
Figure 4 shows the measured data for the 2 mm-wide specimens (obtained from [28]) that are fitted to serve as input for our model:  $R/R_0-\varepsilon$  evolution from a monotonic test (figure 4(a)) and  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$  versus  $\varepsilon_a$  from cyclic tests (figure 4(b)). The correlation between the rate parameters and  $\varepsilon_a$  are reasonably fitted by a power law function, even though different  $\varepsilon_m$  values were used in the 2 mm-wide tests. Figure 5(a) shows the  $\hat{R}_{\max}/R_0$  evolution (in a semi-log format) predicted by the model, using both minimum and initial maximum  $(R/R_0)'$  values, along with the measured  $R_{\max}/R_0$  evolution for selected 2 mm-wide specimens. As expected,  $(R/R_0)'_i$  provides a more conservative  $\hat{R}_{\max}/R_0$  evolution over the cycles than  $(R/R_0)'_{\min}$ . The difference between both modeled curves is more significant for lower  $\varepsilon_a$  values (see for example the  $\hat{R}_{\max}/R_0$  curves for  $\varepsilon_a = 2\%$  vs  $18\%$ ), given the larger differences between  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$  in the low  $\varepsilon_a$  regime (see figure 4(b)). Overall, the  $\hat{R}_{\max}/R_0$  curves using  $(R/R_0)'_{\min}$  are closer to the measured  $R_{\max}/R_0$  evolution than that using  $(R/R_0)'_i$ . These curves are more accurate for the larger  $\varepsilon_a$  values ( $>5\%$ ). For  $\varepsilon_a = 2\%$ , the model is reasonably accurate for the first 1000 cycles, after which the model departs significantly from the measured  $R_{\max}/R_0$  evolution. The inaccuracy for  $\varepsilon_a = 2\%$  is due to the overestimation of  $(R/R_0)'_{\min}$ , given the large spread of  $(R/R_0)'$  measured after 1000 cycles with these low  $\varepsilon_a$  tests (see figure 3(f)).

Figure 5(b) compares the modeled  $\varepsilon_a-\hat{N}_f$  curves for three different  $\varepsilon_m$  values, using both  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$ , to the measured  $N_f$  for 2 mm-wide specimens (obtained from [28]). The modeled curves are nearly identical for  $\varepsilon_m = 15\%$  and  $30\%$ , capturing the fact that  $\varepsilon_m$  has a secondary effect on  $N_f$ . The effect of  $\varepsilon_m$  is non-negligible only for large  $\varepsilon_m$  ( $60\%$ ) and  $\varepsilon_a$  values ( $>10\%$ ) values, as also highlighted by the two fatigue tests at  $\varepsilon_a = 15\%$  ( $N_f = 55$  cycles for  $\varepsilon_m = 30\%$ , and 22 cycles for  $\varepsilon_m = 65\%$ ). Overall, the modeled  $\varepsilon_a-\hat{N}_f$  curves using  $(R/R_0)'_{\min}$  are consistently closer to the measured  $N_f$  than those using  $(R/R_0)'_i$ , which are more conservative (especially for  $\varepsilon_a \leq 3\%$ ).

#### 3.2. Effect of trace width (1 vs 2 mm) on fatigue properties and model

The trace width effect on the electrical behavior of the PE 874 ink has been investigated previously, mainly for monotonic loading [29]. The size effect was related to the crack size being commensurate with the trace widths, the same crack pattern having a more detrimental effect on resistance for smaller trace widths. As such, size effects are not expected for trace widths larger than 2 mm, and the input curves shown in figure 4 could be used to model the fatigue behavior of wide traces. However, the relationship between trace width and fatigue behavior requires additional characterization for narrower traces. Figure 6 shows the  $R/R_0-\varepsilon$  evolution curve and the  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$  vs  $\varepsilon_a$  curves for 1 mm-wide specimens, along with the respective curves for 2 mm-wide specimens. All 1 mm-wide specimens were tested with the same  $\varepsilon_m = 30\%$  (the 2 mm-wide data are from [28], where a range of  $\varepsilon_m$  was used). These figures highlight the extent of trace width effect on the input parameters for our resistance evolution model: the initial resistance (at cycle 1) at the maximum applied strain is increased by up to  $\sim 50\%$ – $60\%$  for the 1 mm-wide specimens (see figure 6(a)), and the rates  $(R/R_0)'$  are increased by a factor of up to 2, especially for larger  $\varepsilon_a$  values (see figure 6(b)). For example, for  $\varepsilon_a = 18\%$  and  $\varepsilon_m = 30\%$ ,  $(R/R_0)'_i$  is 3.22 for 2 mm and 6.11 for 1 mm. The input curves are therefore not solely ink-specific but instead ink- and geometry-specific. This is because the resistance evolution is related to crack evolution [28], and the effect of cracking on resistance is dependent on trace width. The 2 mm data can be used for wider traces, but not but narrower traces.

The trace width effect on the measured  $R_{\max}/R_0$  evolution and the models is illustrated in figure 7(a) for a fatigue test at  $\varepsilon_m = 30\%$  and  $\varepsilon_a = 15\%$ . The experimental curves highlight that the main effect is related to the initial  $R/R_0$  value (for cycle 1), which is twice larger for 1 mm (see figure 6(a)). The figure also shows that the models are much more accurate when using the input data from the correct specimen width. The same conclusion applies for the  $\varepsilon_a-N_f$

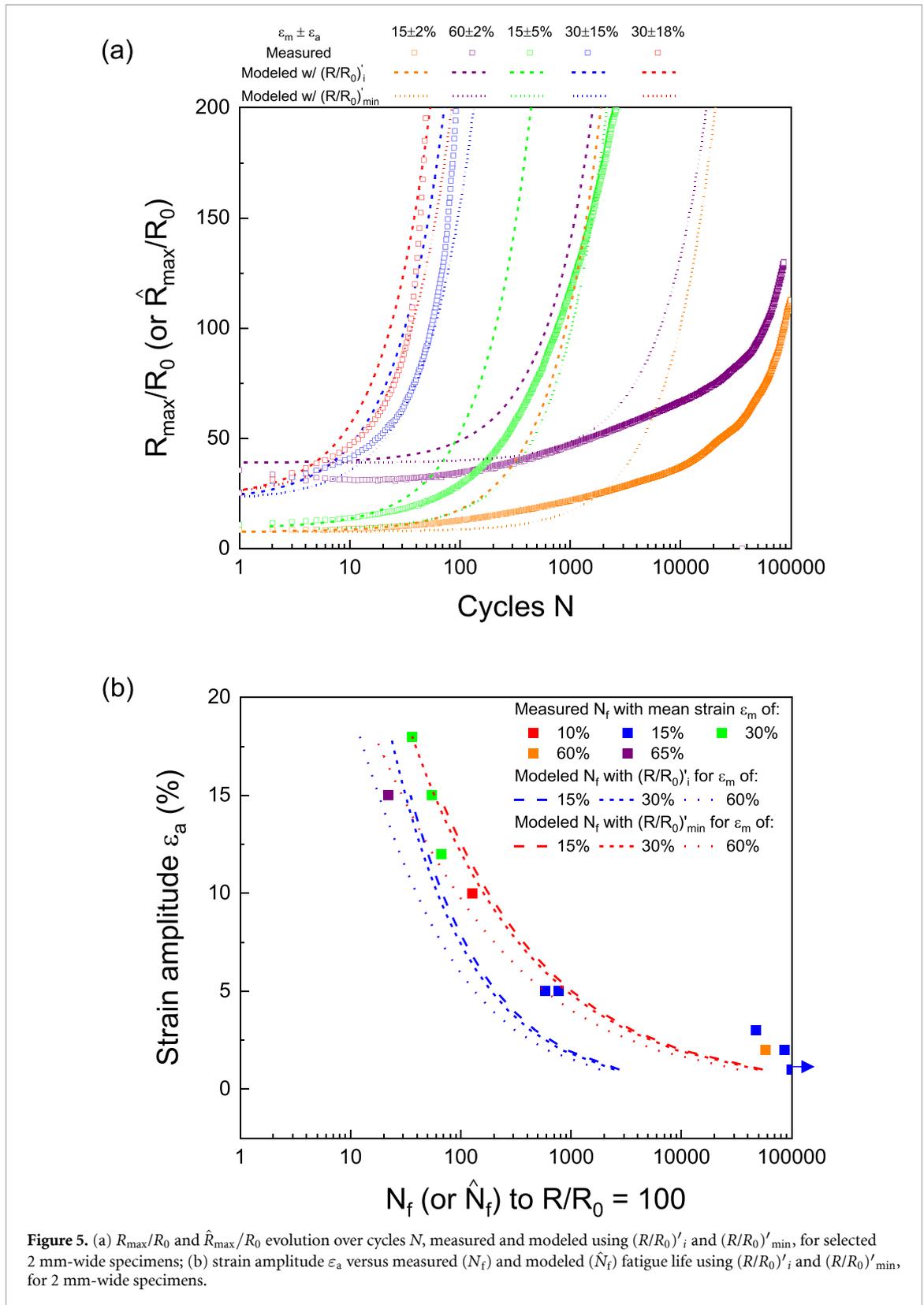


**Figure 4.** Input specimen-characteristic curves for the model. (a) Polynomial fit of experimental data of  $R/R_0-\varepsilon$  for 2 mm-wide-specimens (monotonic test); (b)  $(R/R_0)'_i$  and  $(R/R_0)'_{min}$  for 2 mm-wide specimens as a function of  $\varepsilon_a$ . The solid lines show the power law fit.

curves; see figure 7(b) ( $\varepsilon_m = 30\%$  for the 1 mm data; for the 2 mm data, only  $\varepsilon_m$  in the 15%–30% range is shown, as the effects are negligible in that range). The measured  $N_f$  is consistently lower for the 1 mm-wide specimens, which is why the modeling is trace width-dependent (for trace widths less than 2 mm). The modeled  $\hat{N}_f$  is about twice lower at  $\varepsilon_a = 15\%$  for 1 mm.

**3.3. Determining normalized resistance increase rates using interrupted cyclic tests**

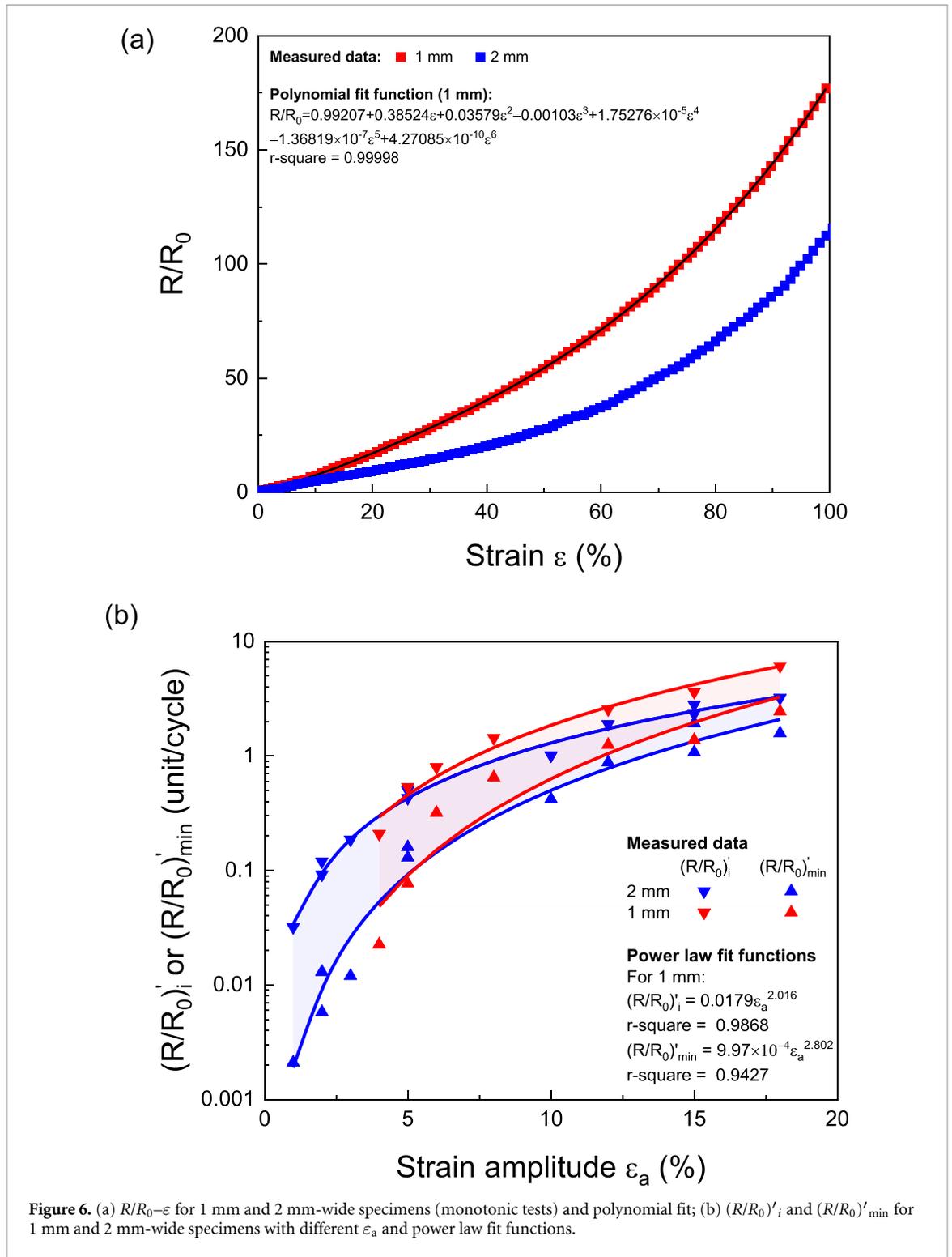
The  $(R/R_0)'-\varepsilon_a$  curves shown in figures 4(b) and 6(b) were obtained by using as many specimens as datapoints (1 datapoint per specimen); see examples in figure 3. The rate curves obtained for 1 mm-wide specimens (see figure 6(b)) are compared to curves obtained from an interrupted cyclic test (see



**Figure 5.** (a)  $R_{\max}/R_0$  and  $\hat{R}_{\max}/R_0$  evolution over cycles  $N$ , measured and modeled using  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$ , for selected 2 mm-wide specimens; (b) strain amplitude  $\epsilon_a$  versus measured ( $N_f$ ) and modeled ( $\hat{N}_f$ ) fatigue life using  $(R/R_0)'_i$  and  $(R/R_0)'_{\min}$ , for 2 mm-wide specimens.

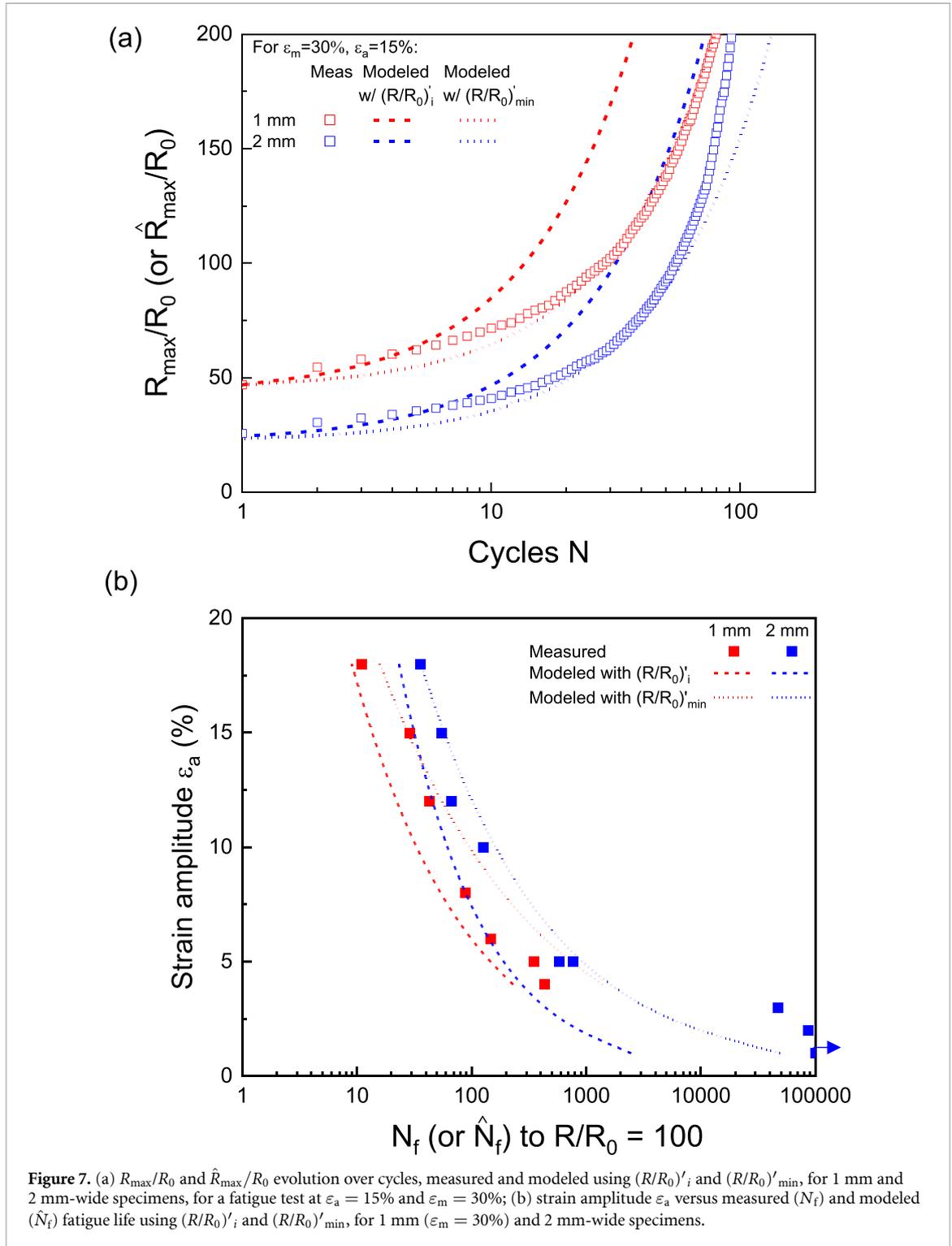
figures 2(e), (f) and section 2.1.2), in order to assess whether the required input data for the model can be obtained with as few experiments as possible. The results of an interrupted cyclic test are shown in figure S1 of the supporting information. For most  $\epsilon_a$  (2%, 4%, 12%, and 15%), there is a decrease in  $R_{\max}/R_0$  between the first and second cycle, after

which  $R_{\max}/R_0$  increases with cycling. This initial value is not included in the calculations, as it is likely related to polymer substrate relaxation due to the unloading/reloading between each  $\epsilon_a$  used for this method. Figure 8 compares the  $(R/R_0)'$  evolution over cycles from the uninterrupted tests (figure 8(a)) and the interrupted test (figure 8(b), starting at  $N = 2$ ).



The main difference between the two methods is the absence of initial decrease in rates for the interrupted test. As summarized in section 2.1.3, for the uninterrupted tests,  $(R/R_0)'$  initially decreases with cycling for all  $\epsilon_a$  values (for  $\epsilon_a = 1\%$  or  $2\%$ , there is initially a decrease in  $R_{max}/R_0$  within the first 30 cycles, before a local maximum initial value in  $(R/R_0)'$  is reached). This difference highlights that the initial decrease in  $(R/R_0)'$  is not an intrinsic property of the specimen, but instead an artifact associated with testing an ink

with no prior loading history. Additional interrupted tests (not shown here) did have an initial decrease in  $(R/R_0)'$  for the first  $\epsilon_a$  value (say,  $\epsilon_a = 4\%$ ), but not for the following  $\epsilon_a$  values, indicating that the initial decrease in  $(R/R_0)'$  only occurs if the ink had no prior loading. The reason for this behavior is unclear. One possibility is the presence of residual tensile stress in the ink. From a practical viewpoint, the model using  $(R/R_0)'_i$  values should not be used to estimate the fatigue behavior of an ink with prior loading history.



**Figure 7.** (a)  $R_{max}/R_0$  and  $\hat{R}_{max}/R_0$  evolution over cycles, measured and modeled using  $(R/R_0)'_i$  and  $(R/R_0)'_{min}$ , for 1 mm and 2 mm-wide specimens, for a fatigue test at  $\epsilon_a = 15\%$  and  $\epsilon_m = 30\%$ ; (b) strain amplitude  $\epsilon_a$  versus measured ( $N_f$ ) and modeled ( $\hat{N}_f$ ) fatigue life using  $(R/R_0)'_i$  and  $(R/R_0)'_{min}$ , for 1 mm ( $\epsilon_m = 30\%$ ) and 2 mm-wide specimens.

Figure 9(a) compares the  $(R/R_0)'_{min}$  obtained from the interrupted test and the uninterrupted tests. The  $(R/R_0)'_{min}$  from the interrupted test are in close agreement with the  $(R/R_0)'_{min}$  obtained from the uninterrupted tests. A power law function was also used to fit the  $(R/R_0)'_{min}-\epsilon_a$  data for the interrupted test. The fact that the two fitting functions for  $(R/R_0)'_{min}-\epsilon_a$ , obtained from interrupted and uninterrupted tests, are similar is remarkable. It implies that reasonable fatigue estimates can be obtained

using the interrupted test, which requires only one specimen (as opposed to seven specimens for the uninterrupted tests). It also implies that the minimum rate is not affected by prior loading history. Li *et al* demonstrated that the normalized resistance increase during fatigue is related to simultaneous widening and deepening of cracks. Therefore, it is likely that prior loading does not affect the way crack widening and deepening occurs at a given  $\epsilon_a$ , and it effects on  $(R/R_0)$ . Figure 9(b) compares the

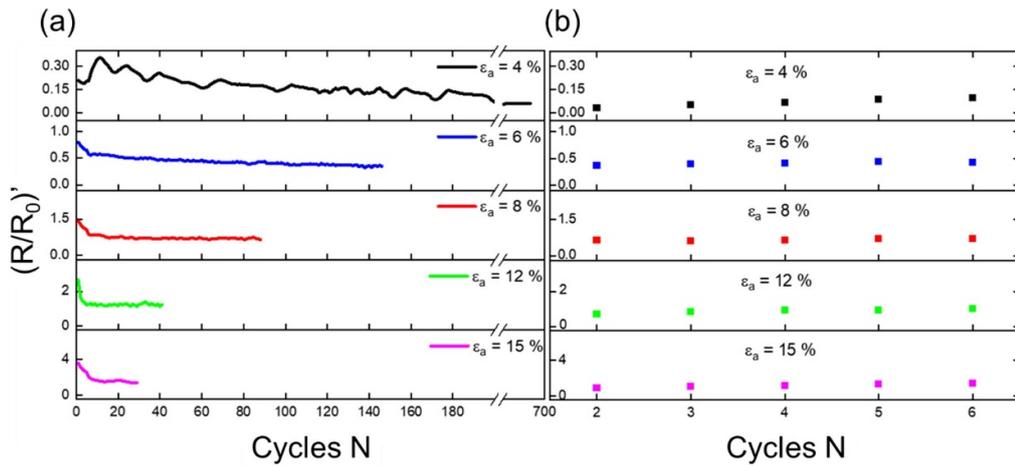


Figure 8.  $(R/R_0)'$  evolution over cycles for (a) uninterrupted tests and (b) interrupted test with 1 mm-wide specimens at  $\epsilon_m$  of 30% and different  $\epsilon_a$  (4%, 6%, 8%, 12%, 15%).

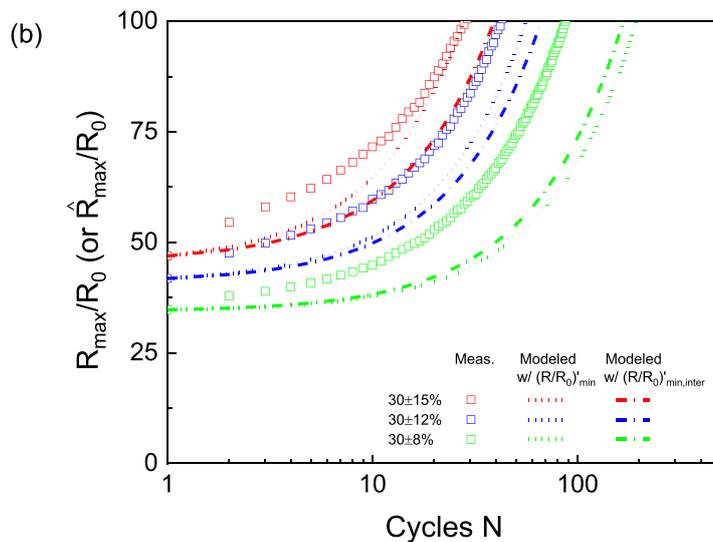
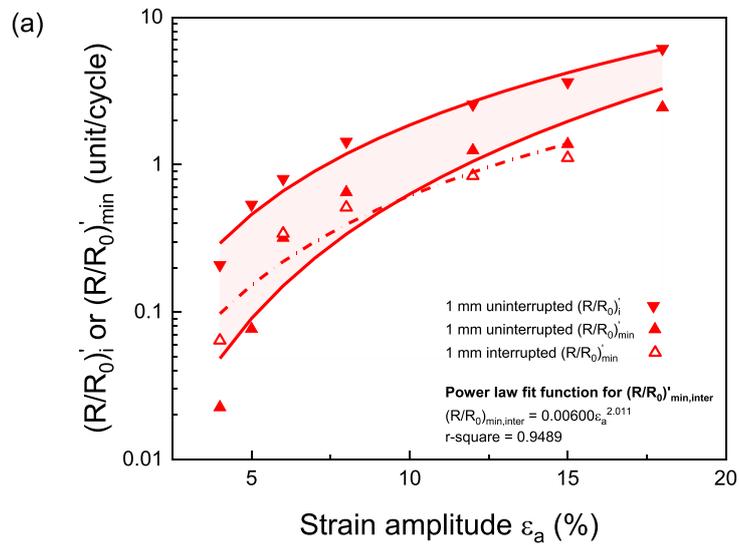


Figure 9. (a)  $(R/R_0)'$  parameters for 1 mm-wide specimens using interrupted and uninterrupted tests with different strain amplitudes  $\epsilon_a$  and power law fit functions for  $(R/R_0)'$  parameters and  $(R/R_0)'$  parameters. (b)  $R_{max}/R_0$  and  $\hat{R}_{max}/R_0$  evolution over cycles  $N$ , measured and modeled using interrupted and uninterrupted curves, for two 1 mm-wide specimens.

modeled  $\hat{R}_{\max}/R_0$  evolution based on the interrupted test fitting function, with the measured  $R_{\max}/R_0$  for three fatigue tests ( $\varepsilon_m = 30\%$ ,  $\varepsilon_a = 8\%$ ,  $12\%$ , and  $15\%$ ). The figure also shows the models based on  $(R/R_0)'_{\min}$  from the uninterrupted tests. Both models provide very similar  $\hat{R}_{\max}/R_0$  evolution, and are in good agreement with the experimental data.

#### 4. Conclusions

The following is a summary of results regarding the formulation and implementation of a rate-based model for predicting electrical resistance evolution and fatigue life of the conductive ink PE 874 under cyclic strain.

- A model to estimate  $R/R_0$  evolution with cycling as well as fatigue life  $N_f$  (as defined by the number of cycles to reach a threshold value of  $R/R_0$ , such as 100) was constructed by using two input specimen-characteristic curves:  $R/R_0$  vs  $\varepsilon$  from a monotonic stretch test, and either  $(R/R_0)'_{i-\varepsilon_a}$  or  $(R/R_0)'_{\min-\varepsilon_a}$  from a set of cyclic stretch tests. The model assumes a constant rate of normalized resistance increase, and incorporates the effects of both  $\varepsilon_m$  and  $\varepsilon_a$ .
- The model predicts the  $R_{\max}/R_0$  evolution for a cyclic stretch test with strain amplitude  $\varepsilon_a$  and mean strain  $\varepsilon_m$  with reasonable accuracy, especially when using the  $(R/R_0)'_{\min-\varepsilon_a}$  input curve. The  $(R/R_0)'_{i-\varepsilon_a}$  curve provides a more conservative estimate but is inaccurate at low  $\varepsilon_a$  ( $<5\%$ ). The mean strain effects are mainly secondary compared to strain amplitude, except for large  $\varepsilon_a$  ( $>10\%$ ) and large  $\varepsilon_m$  ( $>30\%$ ).
- A trace width effect is found for the fatigue behavior of 1 mm vs 2 mm-wide specimens. The input specimen-characteristic curves are trace-width dependent, and the model predicts a decrease in  $N_f$  by a factor of up to 2 for the narrower trace width, in agreement with the experimental results.
- Two different methods were investigated to generate the  $(R/R_0)'_{-\varepsilon_a}$  curves: uninterrupted fatigue tests (requiring  $\sim 6\text{--}7$  cyclic tests to generate the curve), and a single interrupted cyclic test (requiring only 1 specimen tested at progressively higher  $\varepsilon_a$  to generate the curve). The interrupted test can reproduce the  $(R/R_0)'_{\min-\varepsilon_a}$  curve obtained from the uninterrupted tests with reasonable agreement. However, it cannot reproduce the initial maximum rates  $(R/R_0)'_i$ , since it does not exhibit an initial decrease in rates. The results suggest that the initial decrease in rate only occurs for specimen with no prior loading. The minimum-rate curve is therefore recommended for more accurate fatigue estimates.

Although the current model is specifically for the PE874 ink, it is likely to be applicable for other inks

that exhibit a similar fatigue behavior related to ink fatigue cracking.

#### Data availability statement

The data cannot be made publicly available upon publication due to legal restrictions preventing unrestricted public distribution. The data that support the findings of this study are available upon reasonable request from the authors.

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#### ORCID iDs

Q Li  <https://orcid.org/0000-0003-3574-8230>  
 E Chung  <https://orcid.org/0000-0001-7602-5069>  
 A Antoniou  <https://orcid.org/0000-0002-5270-5438>  
 O Pierron  <https://orcid.org/0000-0003-0787-7457>

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