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一种基于组合载荷调控的柔性电子转印方法*

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摘　要　柔性电子器件良好的电学和力学性能突破了传统电子器件的局限性，在仿生电子和医疗检测等领域具有广阔的应用前景. 转印是制备柔性电子的主流技术，实现了将电子器件从制备基体上拾取并印刷至应用基体这一过程. 已有的转印工艺存在印章制备复杂或外部激励引发电子器件损伤等问题，极大地限制了转印的应用范围. 本文提出了一种基于组合载荷调控机制的转印方案，通过控制印章上刚性柱的顺序组合运动，从而调控印章/器件界面位移/应力场，实现界面粘附调控，最后完成在不同刚/柔性基体上的转印. 本文基于理论模型和有限元分析，揭示了转印过程中印章几何参数与能量释放率间的非线性关系，为实际转印印章的设计提供指导性意见. 物理实验证明，本文转印方法不但对电子器件形貌和应用基体具有较高的兼容性，还支持微硅片在柔性基体上的大批量、多层、分次集成.

关键词　柔性电子，转印，组合载荷，粘附调控，界面断裂

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0　引言

柔性电子器件具备轻质、易于携带、能承受大变形、适应于复杂曲面等特征[1,2]，克服了传统电子器件硬、脆的限制，改变了传统电子器件的发展. 在仿生电子、医疗健康检测、能源监测等领域，柔性电子器件具备广阔的应用前景和潜在用途[3-5]，如智能传感蒙皮[6]、软体机器人[7]、仿生电子皮肤[8]、柔性天线[9]、心电图传感器[10]、肌电传感器[11]等. 柔性电子器件制备时常涉及的高温或化学处理等方式限制了其在敏感基体上的直接制备，如人体皮肤等器官. 因此，转印技术在柔性电子器件的制备中扮演了主流的角色[12]，通过印章将电子器件从制备基体上提起并集成印刷在应用基体上[13]. 转印技术将传统电子微纳平面制备与曲面集成解耦，以一种高效、高质量的方式实现柔性电子的多级、多层、多次、异质集成，促进了柔性电子的多样化发展[14].

转印技术主要包括拾取和放置两个过程[15]. 在拾取过程中，需要印章/器件界面的粘性强于器件/制备基体界面的粘性，使得印章能将器件从制备基体上成功提起并转移；在放置过程中，需要印章/器件界面的粘性弱于器件/应用基体界面的粘性，使得器件与印章剥离并被放置在应用基体上. 因此成功转印的关键在于如何实现印章/器件界面强弱粘性调控[16,17]. 研究者们大多通过设计印章和引入外部激励来实现印章/器件界面粘性调控.（1）采用功能材料印章（形状记忆[18,19]、热敏[20,21]、光敏[22]等），利用功能材料独特的应激响应实现印章/器件界面的粘性发生强弱变化；（2）设计印章内部结构（制备空腔[23,24]、引入磁相[25,26]等）或添加表面微结构（浮雕[27]、微柱[28]）等，利用非规则结构的变形调控印章/器件界面的接触面积；（3）利用激光[29,30]、气压[31]等外部激励使印章局部变形导致界面变形不匹配实现界面粘性调控. 虽然已有研究有效实现印章/器件界面粘性强弱转换，但存在功能材料应用范围有限且粘性控制过程相对繁琐、制备不规则印章增加了转印过程的复杂性和转印成本、激光等外部激励易引入残余应力和变形造成印

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章和器件损伤等问题.

为了解决已有方案存在的问题,本文提出一种基于组合载荷调控的通用转印方法. 如图 1 所示,在 PDMS 衬底表面布置多个刚性柱作为转印所需的印章. 在转印过程,通过顺序作动刚性柱,带动 PDMS 衬底与器件形成期望的局部位移/应力场,实现印章/器件间界面粘性调控和成功转印的目的. 本文基于理论模型和数值分析揭示了印章结构设计与界面断裂性能间的关系,为实际印章的设计提供支持. 最后开展了物理实验将多种器件成功印刷集成于不同基体.

1　组合载荷调控转印方法

1.1　工艺流程

图 1 给出本文设计的组合载荷调控的转印流程图. 印章由刚性柱阵列(programmable pillars array)和 PDMS 柔性衬底组成. 在拾取过程中,首先将印章与器件竖直对齐(步骤Ⅰ),对印章施加均匀向下位移场(步骤Ⅱ)使印章的 PDMS 衬底与器件紧密接触,然后对印章施加均匀向上位移场(步骤Ⅲ),利用 PDMS 衬底粘性将器件从制备基体上提起. 在放置过程中,我们需要对刚性柱阵列实现组合载荷调控,为方便起见,当流程图中刚性柱颜色为红色表明对其施加向上位移载荷,刚性柱颜色为黑色则应对其施加向下位移载荷. 如图 1 所示,放置过程中刚性柱的颜色依次由黑色变为红色,代表向上位移载荷(步骤Ⅳ,Ⅴ),与右侧受向下位移载荷的黑色刚性柱间形成较大的界面局部位移差,使得印章/器件界面接触面积和粘附性能降低,实现印章与器件的顺序剥离(步骤Ⅵ),达到在应用基体上集成器件的目的.

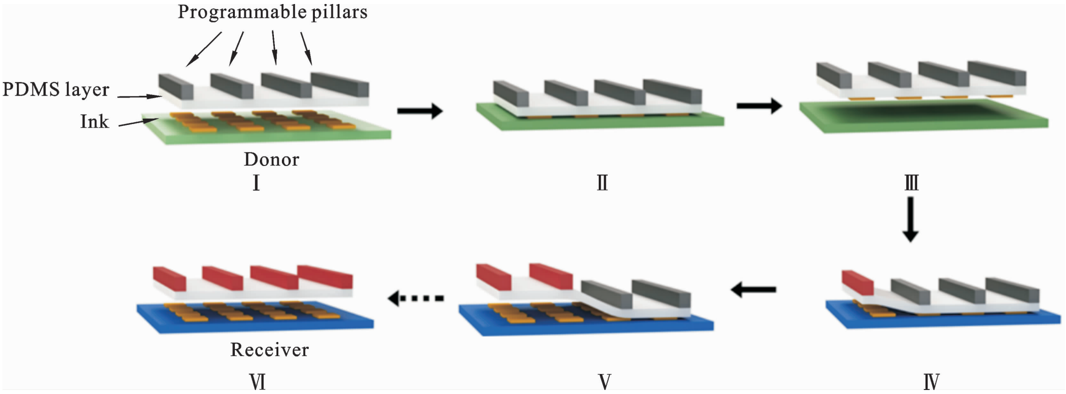


图 1　组合载荷调控转印流程示意图

Fig. 1　Schematic diagram of load-combination modulation transfer printing process

本文中印章由多个刚性柱组成,需要对应的多个作动通道控制刚性柱的独立位移,控制成本高且工艺复杂. 考虑当前刚性柱位移作动具备顺序加载的特点,我们设计了如图 2 所示的转印平台,实现单通道下的刚性柱顺序加载. 转印平台由载荷模块(包括小厚度磁铁 1、大厚度限位块 2 和牵引块 3)和印章(包括刚性柱和 PDMS 衬底)集成于虚线表示的外部支撑框架中,基体置于升降模块上. 载荷模块可在支撑框架内沿轨道做同方向平动,小厚度磁铁可吸引刚性柱发生向上位移,带动连接部分的衬底位移至外部支撑框架中. 在拾取阶段,利用升降模块的向上运动使印章的 PDMS 衬底与器件紧密接触,利用升降模块的向下运动使器件与制备基体剥离. 在放置阶段,利用升降模块的向上运动使器件与应用基体接触,拖动牵引块使得载荷模块在支撑框架中沿轨道方向平动,利用限位块与磁铁厚度差异以及磁铁对刚性柱的吸引作用,实现刚性柱从左到右柱子的依次向上运动,使得柔性衬底层发生弯曲变形,有效降低了印章与器件接触面积,达到与器件脱粘效果,完成器件在应用基体上的集成.

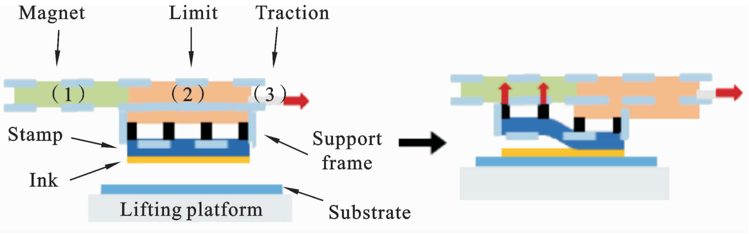
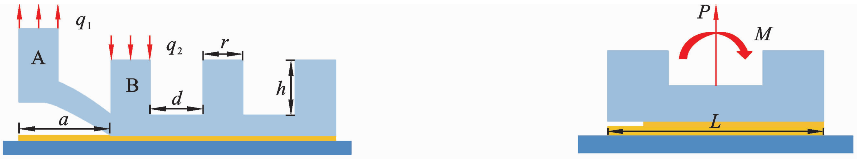


图 2　单通道顺序加载转印平台示意图

Fig. 2　Schematic diagram of transfer printing platform to achieve sequential loads in a single-channel mode

1.2　组合载荷调控转印机理

本文构建理论模型分析组合载荷调控转印的机理，使用 G 表征印章/器件界面裂纹扩展时的能量释放率. 选取放置过程（步骤 1Ⅳ，图 1）为研究对象，截面结构如图 3（a）所示，刚性柱高度为 h，宽度为 r，间距为 d，以柱 A、B 为单胞，如图 3(b)所示，总长为 $L=d+2r$，此时假设印章/器件界面裂纹[32]长度为 a，即 $a=d+r$. 在柱子 A 和 B 顶部分别施加均布载荷 q_1 和 q_2，则单胞模型受等效拉力和弯矩分别为 $P=(q_1-q_2)r$，$M=(q_1+q_2)(d+r)r/2$.



（a）器件放置截面示意图
（a）Sectional illustration of to-be-printed electronic device

（b）受等效拉力和弯矩的单胞模型
（b）Unit cell with the consideration of equivalent tensile forces and bending moments

图 3　转印过程理论模型

Fig. 3　Theoretical model of the transfer printing process

在放置过程中，随着载荷的施加，印章/器件界面开裂. 拉力 P 和弯矩 M 作用下，裂纹尖端的应力强度因子可解析确定为[32]

$$K_{\mathrm{I}}=\sqrt{\frac{\pi a}{\lambda(1-\lambda)^3}}\left(\frac{P\alpha_1}{L}+\frac{M\alpha_2}{L^2}\right) \tag{1}$$

$$K_{\mathrm{II}}=\sqrt{\frac{\pi a}{\lambda(1-\lambda)^3}}\left(\frac{P\beta_1}{L}+\frac{M\beta_2}{L^2}\right) \tag{2}$$

式中 $\lambda=a/L$，$\alpha_1=0.38+0.62\lambda-0.06\mathrm{e}^{-12(\lambda/(1-\lambda))}$，$\alpha_2=2.0-0.72\mathrm{e}^{-9(\lambda/(1-\lambda)}$，$\beta_1=0.17-0.24\lambda-0.02(1-\lambda)^5$ 和 $\beta_2=-0.23+(1-\lambda)^4(0.58-0.2\lambda+0.8\lambda^2)$. 相应地，裂纹尖端的能量释放率 G 为[33]：

$$G=(K_{\mathrm{I}}^2+K_{\mathrm{II}}^2)/(2\tilde{E}) \tag{3}$$

其中系数 1/2 表征印章/器件间材料失配的影响[34]，印章等效弹性模量为 $\tilde{E}\approx E/(1-\nu^2)$，$E$ 为印章杨氏模量，ν 为印章泊松比. 将公式(1)和(2)代入公式(3)可知

$$G=\frac{\pi a(1-\nu^2)[(PL\alpha_1+M\alpha_2)^2+(PL\beta_1+M\beta_2)^2]}{2E(1-\lambda)^3L^4} \tag{4}$$

取 q_2，$l=d+r$ 为固定值，可得无量纲化能量释放率 GE/lq_2^2[35] 与无量纲化参数之间的规律为[36]：

$$\frac{GE}{lq_2^2}=f\left(\frac{r}{l},\frac{d}{l},\frac{q_1}{q_2},\nu\right) \tag{5}$$

从上述公式可知，无量纲化界面能量释放率 GE/lq_2^2 是关于无量纲化参数 $(r/l, d/l, q_1/q_2)$ 的非线性函数. 遵循 Griffth 断裂准则，当裂纹扩展能量释放率大于或等于裂纹扩展所需要的临界能量释放率时，裂纹会发生扩展[13]. 因此能量释放率 G 越大，印章/器件界面越易开裂，实现放置印刷.

1.3　组合载荷调控界面断裂数值仿真

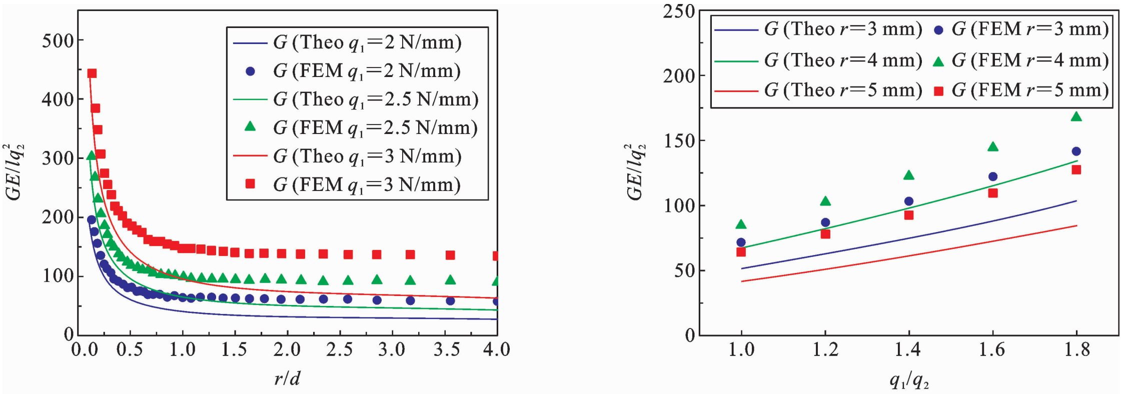
本文利用商用 ABAQUS 软件[37]对单胞模型开展界面断裂力学分析. 有限元分析中刚性柱结构参

数采用宽度 $d=4$ mm，间距 $r=6$ mm，刚性柱材料属性 $E_1=200$ GPa，泊松比 $\nu_1=0.3$；PDMS 衬底层厚度为 4.5 mm，杨氏模量 $E_2=1.5$ MPa，泊松比为 $\nu_2=0.48$；器件厚度为 0.2 mm，杨氏模量 $E_3=190$ GPa，泊松比 $\nu_3=0.278$. 在有限元分析中，设置裂纹长度 $a=10$ mm，固定 $q_2=1$ N/mm，施加不同 q_1 载荷，利用 J 积分[38]求解印章/器件界面能量释放率 G 和评估转印可能性.

2　结果与讨论

2.1　界面断裂性能分析验证

如图 4 所示，我们利用数值仿真验证了公式(5)中界面无量纲化能量释放率 GE/lq_2^2 随柱子宽度与间距比 r/d，载荷比 q_1/q_2 的变化规律. 本文中固定 q_2 为 1 N/mm[39]，q_1 分别取 2 N/mm、2.5 N/mm 和 3 N/mm. 图 4 结果表明，理论计算结果与仿真结果达到较好的吻合程度. 如图 4(a)所示，柱子宽度与间距比 r/d 越大时，无量纲化能量释放率呈现单调下降趋势，说明柱子宽度越小，间距越大，印章/器件界面越容易开裂. 如图 4(b)所示，随着载荷 q_1/q_2 比值增加，无量纲化能量释放率呈现单调上升趋势，说明刚性柱间载荷比值越大，印章/器件界面越容易开裂. 值得注意的是，理论模型采用了半无限体假设[32]，有限元计算则采用与实验条件一致的有限几何结构模型，使得理论解与有限元结果存在一定差异[36]，但两者在关键趋势上一致. 因此，我们仍可利用理论模型为具体转印实验中印章的设计提供重要的理论依据和指导意见，提升转印效果.



（a）无量纲化能量释放率与 r/d 的关系
（a）The relationship between the dimensionless energy release rate and r/d

（b）无量纲化能量释放率与 q_1/q_2 的关系
（b）The relationship between the dimensionless energy release rate and q_1/q_2

图 4　印章结构参数与载荷组合作用下的界面无量纲化能量释放率分析

Fig. 4　The interfacial dimensionless energy release rate for the stamp with different structural parameters and load combinations

2.2　转印实验

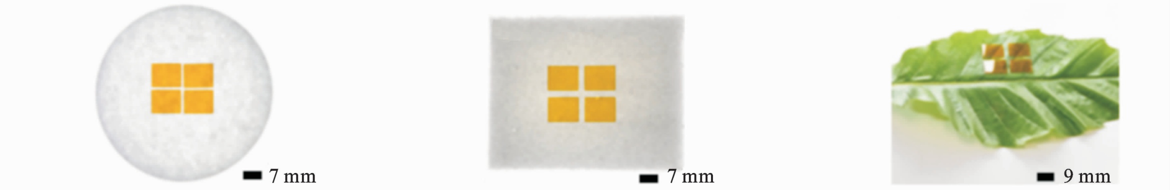
图 5 表明组合载荷调控转印方法对转印器件和基体具备较高的兼容性. 如图 5(a)-5(c)所示，组合载荷调控转印支持不同图案器件，如不同线宽(PI 膜，长 30 mm，厚 100 μm，线宽范围 2～7 mm)、规则多边形(PI 膜，边长分别为 4 mm、10 mm、5 mm 的正六边形、正三角形、五角星)和异形结构(PET 膜，厚 300 μm，DUT 字母图案). 如图 5(d)-5(f)所示，组合载荷调控转印支持不同的基体，如将 2×4 阵列 120 mm×150 mm×100 μm 的 PI 膜分别集成在粗糙基体(滤纸)、多孔基体(泡沫)和脆弱基体(叶子)上.

图 6 表明组合载荷调控转印方法支持大批量、分次和多层转印. 图 6(a)显示了不同尺寸的正方形硅板(最小的为 2 mm×2 mm，最大 8 mm×8 mm，厚度 500 μm)可以一次性转印，表明组合载荷调控转印方法对不同尺寸器件的通用性. 图 6(b)显示组合载荷调控转印可以在已转印成功的两列微硅片(2 mm×2 mm×500 μm，黑色)附近再印刷两列 PI

（a）不同线宽的PI膜
（a）PI films with different line widths

（b）规则多边形PI膜
（b）Regular polygonal PI films

（c）异形PET膜
（c）Irregular PET films



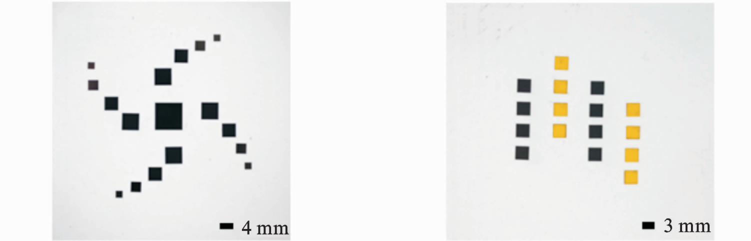
（d）PI膜转印至粗糙基体
（d）PI films printed on rough substrates

（e）PI膜转印至多孔基体
（e）PI films printed on porous matrices

（f）PI膜转印至脆弱基体
（f）PI films printed on fragile substrates

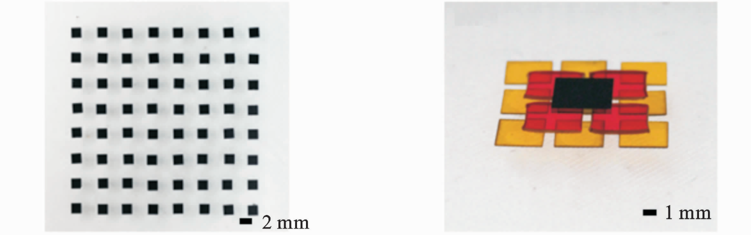
图 5　不同器件在不同基体上的转印

Fig. 5　Transfer printing of different devices on different substrates



（a）转印不同尺寸的微硅片
（a）Transfer printing of micro-silicon wafers with different sizes

（b）分次转印
（b）Multi-time transfer printing



（c）大批量转印微硅片
（c）Large-scale transfer printing of micro-silicon wafers

（d）微硅片与薄膜的多层转印
（d）Micro-silicon transfer printing of micro-silicon wafers and films

图 6　微硅片/薄膜在柔性基体上的分批次、大批量和多层转印

Fig. 6　Multi-time, large-scale and multi-layer transfer printing of micro-silicon wafers/films on flexible substrates

薄膜(2 mm×2 mm×100 μm，黄色)，实现不同材质器件在同一基体上的分批次转印. 图 6(c)显示了 8×8 阵列的微硅片被一次性转印，应用于大规模微器件的高效率集成. 图 6(d)显示了不同材质(PI 膜、PVC 膜、硅片)、不同大小(3 mm×3 mm、4 mm×5 mm、5 mm×5 mm)的器件被依次多层转印，有望实现不同材质的器件多层堆叠和更多元化的功能.

3　结论

本文提出一种简单、普适的组合载荷转印方法

和设计了单通道转印平台，利用刚性柱间组合载荷调控，实现界面粘性调控和成功转印. 理论模型和数值仿真表明，调控刚性柱结构参数和组合载荷差，可有效提高界面开裂能力，保证转印成功率. 转印实验证明本方法适用于不同形状的器件和不同应用基体，以及分批次、大批量、多层转印.

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A Load-Combination Modulation-Based Transfer Printing Method in Flexible Electronics

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Abstract The superior electrical and mechanical properties of flexible electronics enable the breaking of the limitations of traditional electronic devices, and promise wide applications in the fields of bionic electronics, energy monitoring, and medical monitoring. Transfer printing is the mainstream technology for the fabrication of flexible electronics, realizing the processes of picking up electronic devices from the donor substrate and printing them onto the receiver substrate. Transfer printing greatly enriches the fabrication methods of flexible electronics and promotes the development of related industries. However, even with encouraging advantages, current transfer printing processes still face some challenges that cannot be ignored. For example, the preparation process of the stamp is often complex and requires high-precision machining and fine control. In addition, most external excitations cause damage to electronic devices, hindering the further promotion and application of transfer printing technology. To tackle these technical challenges, this paper proposes a load-combination modulation-based transfer printing method. This scheme is proposed to control the loading sequences of rigid pillars on the stamp, modulate the displacement/stress distribution at the stamp/device interface, realize the interface adhesion control, and finally complete the transfer printing on different rigid/flexible substrates. This paper also considers the complex nonlinear relationship between the geometric parameters of the stamp and the energy release rate during the transfer printing process. The related theoretical models and finite element analysis provide valuable insights for the design of actual transfer printing stamps. Physical experiments further validate the effectiveness and reliability of the transfer printing method proposed in this paper. This method not only has high compatibility and adaptability with the morphology of electronic devices and receiver substrates, but also supports large-scale, multi-layer, and multi-time integration of micro-silicon wafers on flexible substrates. Experimental results demonstrate significant application potential and market prospects.

Key words flexible electronics, transfer printing, load-combination, adhesion modulation, interfacial fracture