Self-Assembling Circuits

Plasticity in Self-Assembly: Templating Generates Functionally Different Circuits from a Single Precursor**

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The fabrication of microelectronic circuits now follows a strict, deterministic, top-down plan, and relies on the massive parallelism of photolithography to produce multiple replicates of a device.[1] Neural circuits follow an adaptive, bottom-up plan, and assemble themselves of individual components—cells—following a combination of internal program and external guidance.[2] The work we report herein is an initial step towards a strategy for the fabrication of electrical circuits that also relies on a combination of self-assembly and external guidance. We demonstrate the self-assembly of mm-sized components into 3D aggregates that have structures and patterns of electrical connections determined by the shape of a template—that is, by the geometry of the volume in which the self-assembly proceeds. The components carry light-emitting diodes (LEDs), used as surrogates for more complex microelectronic devices, connected to solder-covered pads. The solder connecting these components—when liquid—acts through capillary interactions to drive the self-assembly of the components, and—when solid—establishes electrical and mechanical connections between selected LEDs. Self-assembly in containers of different shape generates topologically different 3D structures—helices, zigzags, and combinations of the two; these structures have different patterns of electrical connections among the LEDs.

Self-assembly[3,4] based on capillary interactions between solder drops has been used to generate aggregates having a single structure and function.[5] Templating has also been used to bring designed asymmetry to self-assembled aggregates.[6–10] In this work we use templating to control the pattern of electrically functional connections among self-assembled components. Our strategy is to use components that—in the absence of restrictions imposed by a template—self-assemble with equal probability into two different configurations; each of these configurations generates a distinct set of electrical connections between the components. The geometry of the volume available for self-assembly templates the 3D structure of the aggregates, and thus determines the

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patterns of electrical connections formed between the components.

A key issue was to design, and to develop a practical procedure to fabricate, the required components (Figure 1; details of the procedure can be found in the Supplemental Information). Briefly, we defined planar patterns of pads and wires on copper-polyimide composite material by using photolithography (Figure 1 a). We applied manually thermocurable, electrically conducting adhesive to selected pads. Folding of the flexible substrate positioned the copper pads and wires in three dimensions (Figure 1 b, c). Curing the components at a temperature above the glass transition temperature of the polyimide consolidated the four-layered, folded structures into rigid, rhomboid components, and simultaneously solidified the electrically conducting adhesive (Figure 1 d). This strategy for the fabrication of the components allowed us to establish electrical connections in three dimensions by using standard photolithography to pattern planar substrates.[11]

We manually soldered two different LEDs—red and green—onto contact pads on each side of the components, and dip-coated the copper pads with solder ($T_m = 70\,°C$) (Figure 1 e, f). The pairs of identical LEDs on opposite sides of each component were electrically connected in series, and shared a common cathode—that is, the circular central pad δ (Figure 1 g). The components were suspended in dilute aqueous acetic acid (pH 3–4),[12] and heated to a temperature above the melting point of the solder (≈80 °C). Rotation of the vessel containing the components caused them to collide with one another and with the walls of the container; capillary interactions between the drops of molten solder caused self-assembly.[5,13]

Each component could interact with, and connect to, two other components. In previous work on patterned self-assembly, the design of the interacting patterns was focused on achieving unique orientational selectivity during self-assembly.[14,15] Here, the threefold symmetry of the patterns of solder could, in principle, allow for three relative orientations in which all pads of solder carried by two components are fully aligned (Figure 2 a). One of these three orientations,

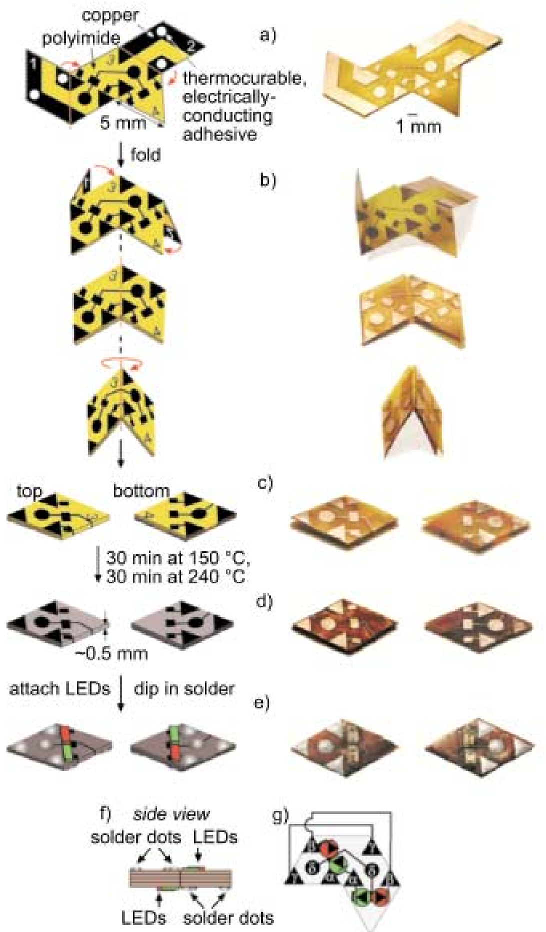

Figure 1. Procedure used for the fabrication of the components. a) Planar patterns of pads and wires were defined on a flexible copper–polyimide composite by using photolithography and wet etching. b) Folding of these patterns resulted in rhomboid structures (c). d) Thermal curing consolidated the four-layer structure. e) LEDs were soldered manually onto the contact pads, and the remaining copper pads were dip-coated with solder. f) Side view of a component after folding and internal interconnection. The thickness is drawn out of scale to emphasize the four-layer structure. g) Scheme describing the electrical connections between pads of solder and LEDs on both sides of one component. The anodes of the green LEDs were connected to pads α, and those of the red LEDs to pads β. Electrical connections (shown as thick solid lines) between symmetrical pads on both sides (β–β and γ–γ) were established by the electrically conducting adhesive.

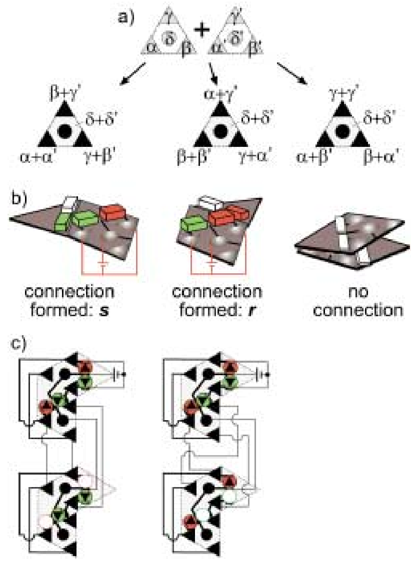

Figure 2. Configuration of the components in an aggregate and pattern of electrical connections formed between the LEDs. a) Two patterns of solder pads (indicated with α, β, γ, and δ on one component, and α′, β′, γ′, and δ′ on another one) can, in principle, overlap completely in three indistinguishable orientations. b) When attached to components, the same pattern of solder pads can overlap completely in only two orientations, *r* or *s*. c) Circuit diagrams describing the electrical connections formed between two components assembled in two different relative orientations, *s* and *r*, after connecting to a battery. Thick lines indicate the electrical connections existing prior to binding of the components, and thin lines indicate the connections formed during binding. The LEDs connected in electrical circuit are shown in their respective color (red or green), and those not included in the circuit are shown in white.

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however, was sterically excluded by the interaction of the multilayer plate of one component with the LEDs on the second (Figure 2b, right). The components could interact in two distinct configurations, *r* and *s* (Figure 2b, left and middle). In both configurations, the overlapping circular central pads (δ and δ′) of adjacent components connect the cathodes of all LEDs in an aggregate in series. In configuration *s*, overlapping peripheral pads α and α′ connect in series the anodes of the green LEDs, and in configuration *r*, overlapping peripheral pads β and β′ connect in series the anodes of the red LEDs. Thus, the different relative orientation of the components results in the formation of electrical circuits that include different LEDs (Figure 2c). When two components connected in configuration *s*, on one component both the green and the red LEDs lighted, and on the second one, only the green LEDs lighted; when the components were connected in configuration *r*, on one component both the green and the red LEDs lighted, and on the second one, only the red LEDs lighted.

When we allowed the self-assembly to proceed in a spherical container or in a cylindrical container with a diameter of ≥ 50 mm, both *r* and *s* connections formed with equal probability. The size of the container, and thus the volume available for self-assembly, imposed no geometric constraint on the shape of the aggregates formed; shearing forces favored irregular aggregates with aspect ratio ≈ 1.

When we allowed self-assembly to proceed in a cylindrical container with a diameter of 8 mm and a volume of ≈ 2 mL that rotated along its principal axis, alternating *r* and *s* connections formed between the components (Figure 3a). The resulting aggregates had a zigzag shape and width of 4.5 mm; no other arrangement of components could fit in the restricted volume available.

When we allowed the self-assembly to proceed in a container with a diameter of 12 mm and a volume of ≈ 2.6 mL that rotated about an axis normal to its own, the restricted volume available for self-assembly of the components geometrically excluded the formation of aggregates with randomly mixed *r* and *s* connections. Zigzag aggregates containing alternating *r* and *s* connections had a high aspect ratio and were, thus, susceptible to disruption by shearing and broke. The geometry of the container favored formation of aggregates containing only *r* or only *s* connections, that is, left-handed or right-handed helices (Figure 3b). The two types of helix initially formed with equal probability, but one type of chirality dominated at the conclusion of the assembly.

Aggregates containing both zigzag and helical regions assembled in a container comprising two cylindrical regions of different width (Figure 3c). The components contained in the narrow portion of the container assembled into a zigzag, and the components contained in the wider portion assembled into a helix. The formation of the two aggregates proceeded independently. The two regions of the container were orthogonal to each other, and the two segments eventually combined into a single, asymmetrical aggregate.

When cooled, the solder drops solidified, bridged the components, and provided both mechanical stability and electrical connectivity. After self-assembly, we connected the aggregates to a power source. The pattern of electrical

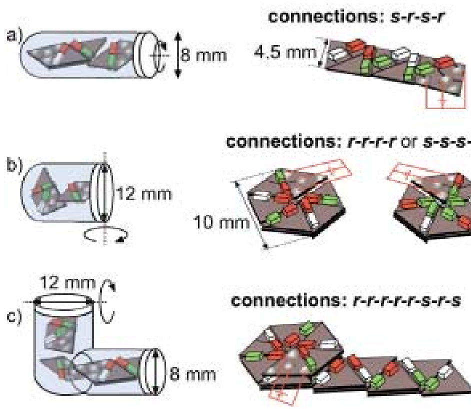

Figure 3. Design of the self-assembling system. The different shapes of the containers in which self-assembly was performed (left) templated formation of aggregates of different geometry (right): a) zigzag, b) left- or right-handed helices, and c) a combination of a zigzag and a helix.

connections that had formed during the assembly was apparent from the pattern of illuminated LEDs. As expected, in aggregates of different geometry the sequence of illuminated LEDs followed different patterns: in zigzag aggregates, (red + green)–(green)–(green)–(red + green)–(red)–(red)–etc. (Figure 4a); in left-handed helical aggregates, (red + green)–(red)–(red + green)–(red)–etc. (Figure 4b); in right-handed helical aggregates, (red + green)–(green)–(red + green)–(green)–etc. (Figure 4c); in aggregates combining helical and zigzag regions, the pattern was a combina-

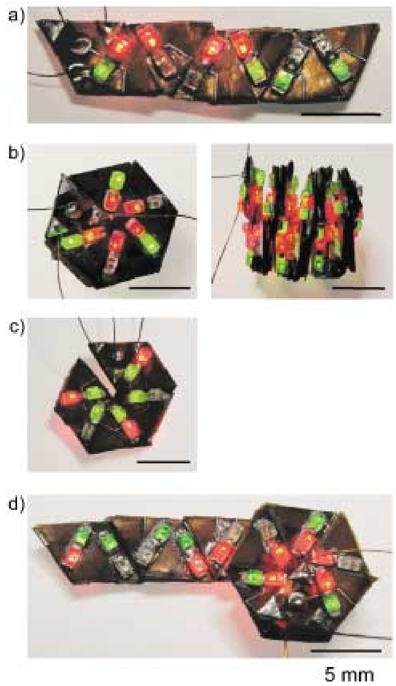

Figure 4. Photographs of self-assembled aggregates shaped as a) a zigzag, b) a left-handed helix shown in top (left) and side (right) views, c) a right-handed helix and d) a combination of a right-handed helix and a zigzag, after connecting to a battery.

tion of the characteristic patterns for a helix and a zigzag (Figure 4d).

This work demonstrates the use of a macroscopic variable—the geometry of the vessel, and thereby the space available for self-assembly—to direct the self-assembly of a single kind of component in distinct patterns of electrical connections. The work establishes that, in principle, multiple functions can be derived from the same set of components by controlling the shape of the space available for self-assembly. Such external means of control over functional connectivity within self-assembled aggregates might be especially important when the components used to build the aggregates are small, and, therefore, difficult to manipulate by using established pick-and-place fabrication strategies.[16]

To achieve a high volumetric density of functional components in self-assembled aggregates, the components must be smaller than those used in this work. The strategy we used for the fabrication of the components—a combination of photolithography and folding—allows (in principle) the scaling of the components to micrometer sizes: conventional 2D photolithography can be used to fabricate and pattern planar precursors, which can then spontaneously fold into 3D shapes.[5,17,18]

The principle of the design that we describe can be used to develop functional, self-assembled systems in which one set of components—carrying functional elements, for example, microelectronic devices—can be combined into any of several functionally different circuits. This strategy reduces the need to fabricate a different set of components for every new pattern of connections. In our experience, the fabrication of the individual components is the most difficult problem when designing functional, self-assembling systems.

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