

Stretchable GaAs Photovoltaics with Designs That Enable High Areal Coverage

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Recent research in advanced materials and mechanics demonstrates the possibility for integrating inorganic semiconductors with soft, elastomeric substrates to yield systems with linear elastic mechanical responses to strains that significantly exceed those associated with fracture limits of the constituent materials (e.g. ~1% for many inorganics). This outcome can provide stretching to strain levels of tens of percent (in extreme cases, more than 100%), for diverse, reversible modes of deformation, including bending, twisting, stretching or compressing.^[1–7] Interest in these outcomes is motivated by needs in flexible display,^[8–10] curvilinear imaging devices,^[11–13] structural health monitors^[14] and, more recently, in bio-integrated systems^[15–17] for advanced therapeutic or diagnostic functionality in clinical medicine. In these latter applications, considerations related to toxicity and biocompatibility of the materials are also critically important. Some of the most well developed strategies exploit configurations in which brittle, rigid materials accommodate

in-plane strains through out-of-plane motions, via buckling or twisting modes.^[18] Such ideas can be exploited in all parts of an integrated system, or only in interconnections between active devices. The latter design can accommodate the largest strains, but its efficacy decreases as the areal coverage of the devices increases.^[12] As a result, important applications such as those in light capture (i.e., photovoltaics) and detection (i.e., photodetectors), where high coverages are often desired, can be difficult to address. Here we report designs for stretchable systems that exploit elastomeric substrates with surface relief that confines strains at the locations of the interconnections, and away from the devices. The results enable areal coverages and levels of stretchability with relatively low interfacial stress between devices and substrates, compared to similar layouts with conventional, flat substrates. We describe, using a combination of theory and experiment, the essential mechanics, and then demonstrate the ideas in stretchable solar modules that use ultrathin, single junction GaAs solar cells.

A representative layout for a structured substrate designed for this purpose appears in **Figure 1**, in which the relief consists of isolated, raised regions (i.e. islands) separated by recessed features (i.e. trenches). The casting and curing processes of soft lithography^[19] provide a convenient means to form such relief, with excellent dimensional control, in elastomers such as poly(dimethylsiloxane) (PDMS). The image of **Figure 1a** provides a cross sectional view for a representative case where square islands with edge lengths (l_{island}) of 800 μm are separated by trenches with widths (l_{trench}) and depths (h_{trench}) of 156 μm and 200 μm , respectively. The thickness of the underlying PDMS (i.e. base) is 200 μm . This type of structure is attractive for stretchable systems that incorporate non-stretchable active elements at the islands because it isolates strains from these regions. In particular, for this example, stretching the substrate (**Figure 1b**) to an overall strain of ~20% induces elongations (~123%) in the trenches that are much higher (>300 times) than those at the islands (~0.4%). The finite element method (FEM) quantitatively captures this basic mechanics, as shown in **Fig 1b**. In particular, for 20% overall strain, FEM yields a 124% increase in l_{trench} (i.e. 156 μm to 349 μm), which is very close to the value (~123%) obtained from experiment. FEM gives a very small change (0.3%) in l_{island} , similar to that (~0.4%) from experiments (see Supporting Information).

The resulting mechanics ensures that brittle inorganic devices mounted on the surfaces of the islands do not experience substantial mechanical stresses upon stretching or other mechanical deformations such as bending or twisting. This outcome obtains even for substrate layouts that consist primarily

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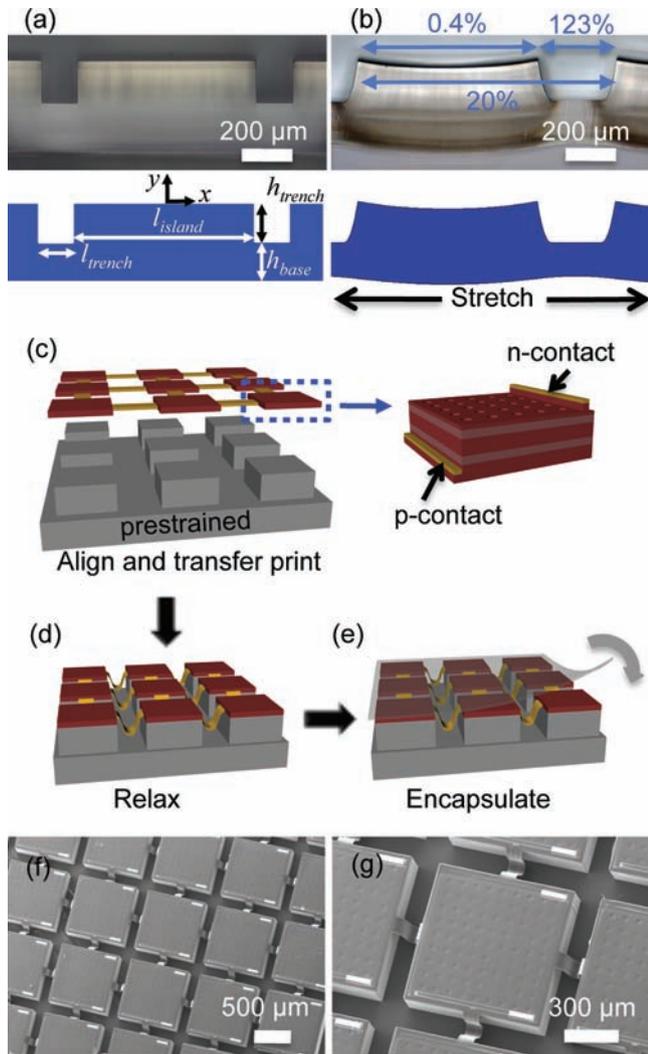


Figure 1. Optical microscopy images and finite element method (FEM) calculations of deformation of a slab of poly(dimethylsiloxane) (PDMS) and schematic illustrations of steps in the fabrication of stretchable GaAs photovoltaic modules with SEM images of a completed device. (a) Cross-sectional optical microscopy images (upper) and FEM (lower) of a slab of PDMS in relaxed (a) and stretched (b) states, with a square array of raised islands (each $\sim 800 \mu\text{m} \times 800 \mu\text{m}$) separated by recessed trenches (widths $\sim 156 \mu\text{m}$, depths $\sim 200 \mu\text{m}$), designed as a platform for stretchable inorganic photovoltaics with high areal coverages. The latter image shows clearly that stretching (in this case, overall applied strain of 20%) induces dimensional changes at the top surfaces of the islands ($\sim 0.4\%$) that are much smaller than those at the trenches ($\sim 123\%$). This mechanics isolates devices located on the islands from applied strain. The FEM results show quantitative agreement with the experimental image in (b). (c) In the first step of the fabrication, an array of ultrathin GaAs microcells is bonded onto a prestrained, structured substrate of PDMS. Each microcell (size $\sim 760 \mu\text{m} \times 760 \mu\text{m}$, thickness $\sim 3.6 \mu\text{m}$) has top (n) and bottom (p) Ohmic contacts that connect to thin electrical interconnect ribbons, in the overall geometry of an open mesh. (d) Releasing the pre-strain deforms the interconnects into arch-shaped bridges, usually in a vertical, upward orientation as illustrated here. Pressing on the entire array using a flat plate of glass causes these bridges to buckle downward, into the trench regions of the PDMS substrate. (e) In a final step, a thin ($\sim 50 \mu\text{m}$), uniform layer of PDMS is bonded on top, as an encapsulant to protect the system from the environment. For purposes of imaging and electrical probing, the modules are left unencapsulated unless specifically noted. (f,g) SEM images of a completed module.

of islands, thereby making it possible to realize areal coverages of devices that are significantly higher than those achievable with conventional, flat substrate designs.^[5–7,10] For photovoltaic modules that consist of interconnected arrays of an ultrathin, GaAs solar microcell technology,^[20–22] the fabrication begins with vertical etching through active layers grown epitaxially on GaAs wafers, to define lateral dimensions of cells with sizes and spacings matched to the islands on the PDMS, in its biaxially prestrained state. Releasing these devices by etching a buried sacrificial layer of ALAs, and then performing further processing on a temporary substrate yields Ohmic contacts and thin ribbon-like type interconnects between adjacent cells, as illustrated in the upper frames of Figure 1c. The inset on the right provides a schematic view of a single cell, with p and n contacts. The interconnects consist of films of gold ($0.32 \mu\text{m}$ thick) coated on top and bottom with layers of an epoxy polymer ($1.0 \mu\text{m}$ and $2.0 \mu\text{m}$ thicknesses on top and bottom, respectively). The GaAs microcells are $3.6 \mu\text{m}$ thick, and they include the same polymer coatings. See Supplemental Information for details. Another transfer printing step delivers the resulting mesh structure onto a prestrained structured PDMS substrate, as in the lower left frame of Figure 1c. Relaxing the strain (Figure 1d) induces compressive forces localized at the trenches, according to the mechanics of Figure 1b. The interconnects respond to this strain by buckling into arc-type shapes, most ($>90\%$) with an upward orientation. Pressing down on the entire system with a flat, rigid plate causes the interconnects to buckle downward, into the trenches as shown in Figure 1d. The final step involves bonding a thin, uniform layer of PDMS on top, as illustrated in Figure 1e, to serve as an encapsulant, by exploiting interface covalent bonds that form between hydroxyl groups on the surface of the PDMS layer and a coating of SiO_2 evaporated on the devices, as described in the Supporting Information. The scanning electron microscopy (SEM) images of Figure 1f,g show a module, with the microcells and buckled interconnects clearly visible. Each microcell includes exposed regions at the p and n contacts to facilitate electrical measurements of individual devices.

Figure 2 shows these kinds of modules, in the relaxed state (Figure 2a), biaxially (Figure 2b) and uniaxially (Figure 2c) stretched, in a complex, twisting deformation (Figure 2d), laminated into sheets of fabric (Figure 2e) and paper (Figure 2f), and extremely bent (Figure 2g). The strains in the interconnect ($\epsilon_{\text{interconnect}}$) and device islands ($\epsilon_{\text{deviceisland}}$) for the as-fabricated state can be given analytically in terms of the overall strain ϵ

$$\epsilon_{\text{interconnect}} = 4\pi\gamma_{\text{interconnect}} \frac{[\epsilon(l_{\text{island}} + l_{\text{trench}})]^{1/2}}{[\epsilon l_{\text{island}} + (1 + \epsilon)l_{\text{trench}}]^{3/2}} \quad (1)$$

$$\epsilon_{\text{deviceisland}} = 4\pi\gamma_{\text{deviceisland}} \frac{[\epsilon(l_{\text{island}} + l_{\text{trench}})]^{1/2}}{[\epsilon l_{\text{island}} + (1 + \epsilon)l_{\text{trench}}]^{3/2}} \times \frac{(EI)_{\text{interconnect}}}{(EI)_{\text{deviceisland}}} \quad (2)$$

where $\gamma_{\text{interconnect}}$ and $\gamma_{\text{deviceisland}}$ are the distances to the neutral mechanical plane of the interconnect and device islands, respectively, and EI is the corresponding bending stiffness (see Supporting Information for details). For

$\varepsilon = 20\%$, the maximum strains in gold (interconnect) and GaAs (device islands) are 0.94% and 0.22%, respectively. Consistent with expectation based on the mechanics of Figure 1b, uniaxial stretching by 20% induces 123% elongation in the trenches whose lengths lie perpendicular to the applied strain. Simultaneously, the trenches in the orthogonal direction narrow by 44%, due to the Poisson effect, which agrees well with the value (46%) from FEM, and can be estimated analytically (see Supporting Information for details)

$$\text{contraction} = \frac{\nu\varepsilon}{\frac{l_{\text{trench}}}{l_{\text{island}} + l_{\text{trench}}} \left(1 + \frac{l_{\text{trench}}}{l_{\text{island}} + l_{\text{trench}}} \frac{h_{\text{trench}}}{h_{\text{base}}}\right)} \quad (3)$$

where ν is the Poisson's ratio. This 44% contraction is equivalent to an overall strain of $\varepsilon = -7.2\%$, as obtained from $\varepsilon (l_{\text{island}} + l_{\text{trench}}) / l_{\text{trench}} = -44\%$. The corresponding maximum

strains in the gold interconnects and the GaAs are obtained from Equation (1) and (2) as 1.1% and 0.25%, respectively. The maximum strains in the polymer at the interconnects and device islands are 6.2% and 0.51%, respectively. Slight design modifications can reduce the strain in the interconnects. For example, by fixing the thicknesses of gold and polymer, but shifting the gold toward the neutral mechanical plane, the maximum strains in the gold and polymer can be reduced to 0.51% and 5.2%, respectively. Failures at large strain deformations are typically due to fracture of the metal layers in the interconnects. Further reductions can be achieved using interconnects with curved shapes.^[5] Even with the relatively simple implementation here, the modules can be twisted into complex shapes (Figure 2d) without any damage to the devices or interconnects. These favorable mechanical properties also allow them to be integrated onto unusual substrates such as cloth (Figure 2e) and paper (Figure 2f). The module in Figure 2e is

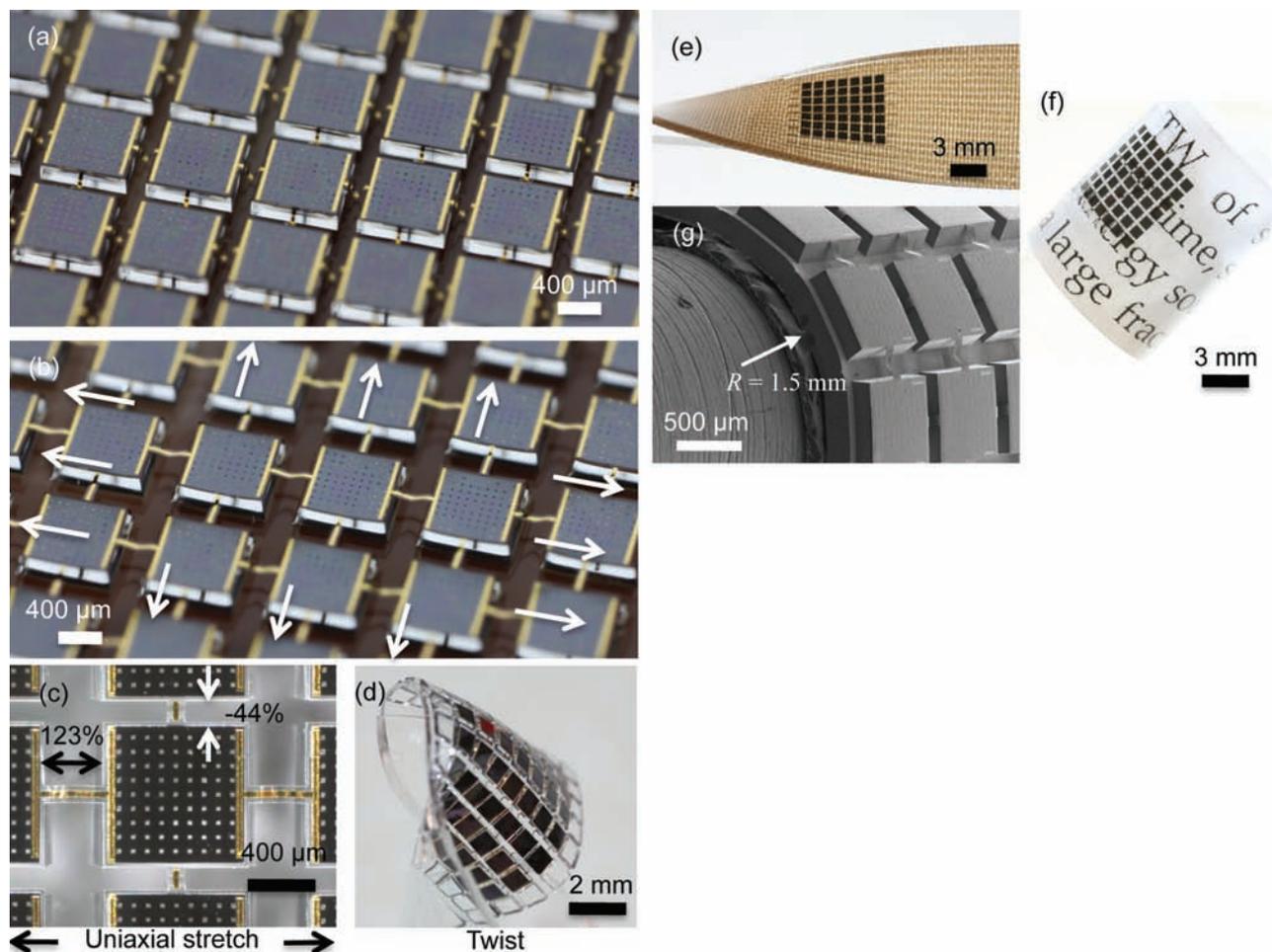


Figure 2. Optical microscope and SEM images of stretchable GaAs photovoltaic modules in various states of deformation and integrated onto different substrates. (a) Representative module, with a ~70% areal coverage of islands. (b) Similar module, in a state of 20% biaxial strain. Here, the regions of the trenches and islands stretch by ~123% and a negligible amount, respectively. This mechanics isolates the GaAs microcells from applied strain. (c) Module in a state of 20% uniaxial strain. Here, the widths of the trenches narrow (~44%) in the direction perpendicular to the stretching direction, due to the Poisson effect. In both directions, the dimensions of the islands change by a negligible amount. Quantitative analysis of the strain appears in the supplementary information. (d) Stretchable GaAs photovoltaic twisted into a complex shape, to illustrate the level of deformability. Similar module, laminated on a piece of cloth (e), twisted 90° over the length of 24 mm and paper (f), wrapped on a cylinder with a radius of 5.8 mm. (g) SEM image of the module wrapped on a cylinder with a radius of 1.5 mm.

twisted 90° over the length of 24 mm. The module in Figure 2f is wrapped on a cylinder with a radius of 5.8 mm.

A simple model predicts the minimum bending radius by assuming fracture upon further bending after fully straightening the interconnects. See Supporting Information for details. The calculated minimum bending radius is 1.4 mm. Experimental results in Figure 2g indicate that the module can wrap a cylinder with a radius of 1.5 mm with a little room to bend further as the interconnects are not fully straightened at the curvature. The island-trench design also maintains a flat geometry for the device islands, consistent with low strains at these locations, even at the extreme bending as shown in the SEM image in Figure 2(g). The trenches absorb the strain caused by bending the module. In this case, the calculated minimum bending radius of the module is 11.2 mm for the opposite direction, if we assume that failure occurs when the strain in the Au reaches 1%. With a higher prestrain (e.g. ~30%) and correspondingly longer interconnects, we can achieve, at the same as-fabricated areal coverage of ~70%, bending radii of 1.8 mm in both directions as described in Supporting Information.

Quantitative study of the mechanics by modeling and experiment defines the range of deformations that can be accommodated for different designs, including aspects such as the Poisson effects mentioned above and all others. Furthermore, the results allow comparison of the strategies reported here to systems based on conventional, flat PDMS substrates. The sizes of the devices, their spacings and other parameters are the same for both cases, as given in Figure 1, as are the total thicknesses ($h_{\text{base}} + h_{\text{trench}}$ for the structured case). See Figure 3a and b. A key difference is that a biaxial prestrain of 20% gives a biaxial stretchability of 20% for the structured case; the flat substrate requires a larger prestrain to achieve the same stretchability. This behavior occurs because the prestrain stretches a flat substrate uniformly, but this strain does not fully relax in the regions of the devices due to their mechanical loading. In particular, analysis by FEM suggests that a biaxial prestrain of 30.0% is needed to achieve 20% stretchability in the flat case (see Supporting Information for details). Figure 3c–f show the distributions of shear and normal stresses at the interface between the microcells, for structured and flat substrates, respectively. The maximum shear and normal stresses for the structured substrate are about 8 and 7 times lower than those for the flat substrate, respectively. The structured geometry therefore offers a significant advantage against shear mode failures at the interface. Another difference between the structured and flat substrates, immediately observable in systems with high areal coverages, relates to overall bowing effects, similar to those previously explored by us in the context of uniform, rigid thin layers on soft substrates.^[23] As illustrated in optical images and FEM results of Figure 3g,h, with the parameters and prestrains described previously, modules on flat substrates exhibit strong bowing, due to high interfacial stresses.

To demonstrate these devices and mechanics considerations in functioning modules, we evaluated the photovoltaic performance in relaxed (Figure 4a) and stretched (Figure 4b) states under using a solar simulator (Oriel, 91192) as shown in Figure 4c. The open circuit voltage and short circuit current of an individual microcell are 0.91 V and 88 μA respectively, as shown with the

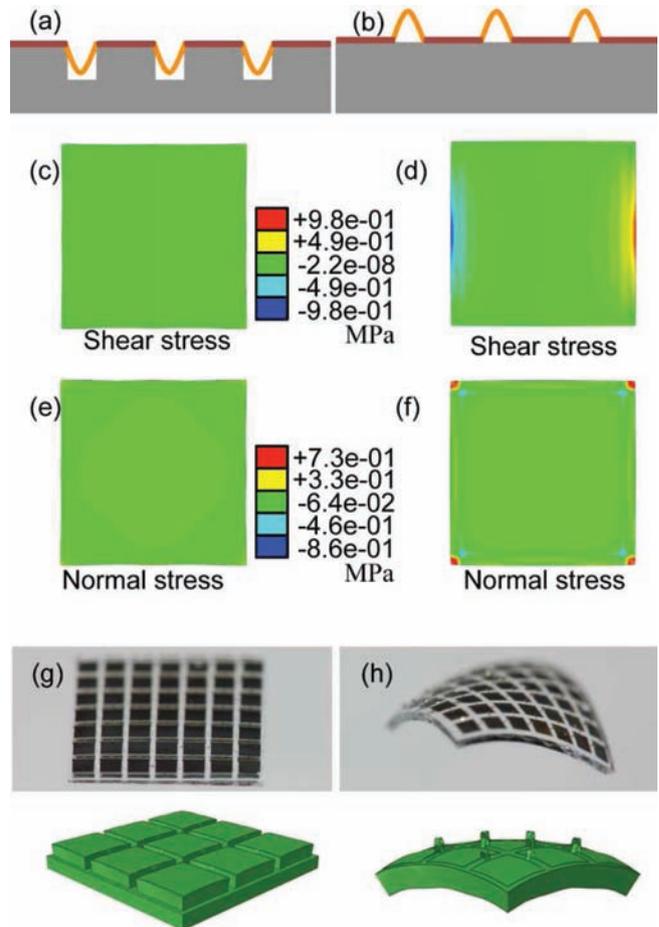


Figure 3. Finite element method (FEM) results for stretchable GaAs photovoltaic modules, consisting of interconnected arrays of devices in non-coplanar, mesh geometries, bonded to structured and flat elastomeric substrates. The islands (~800 $\mu\text{m} \times 800 \mu\text{m}$) cover 70% of the structured substrate. The widths and depths of the trenches are 156 μm and 200 μm , respectively. The total thicknesses of the structured and flat substrate are both 400 μm . Schematic illustrations of the structured (a) and flat (b) cases. FEM results for the interfacial shear stresses between the devices and the structured (c) and flat (d) substrates. The maximum shear stress for structured substrate is eight times lower than this for the flat substrate. FEM result for the interfacial normal stresses between the devices and the structured (e) and flat (f) substrates. The maximum normal stress for the structured substrate is seven times lower than this for the flat substrate. (g) Bowing effects observed by experiments (upper) and FEM (lower) for devices on structured (g) and flat (h) substrates. For substrates with these thicknesses, bowing is pronounced in the flat case but not the structured case, due to comparatively high interfacial stresses between the devices and substrate for the former.

blue curve. Energy conversion efficiencies and fill factors are approximately 13% and 0.79, respectively, for each microcell. As expected, the open circuit voltage increases about seven times (~6.4 V) with seven microcells connected in series, in the relaxed state (Figure 4c, black curve). Biaxial stretching to 20% (red curve in Figure 4c) does not change these characteristics, to within experimental uncertainties. Furthermore, cycling tests involving repeated biaxial stretching to 20% followed by complete relaxation leave the performance unchanged. The efficiency (~12.5%)

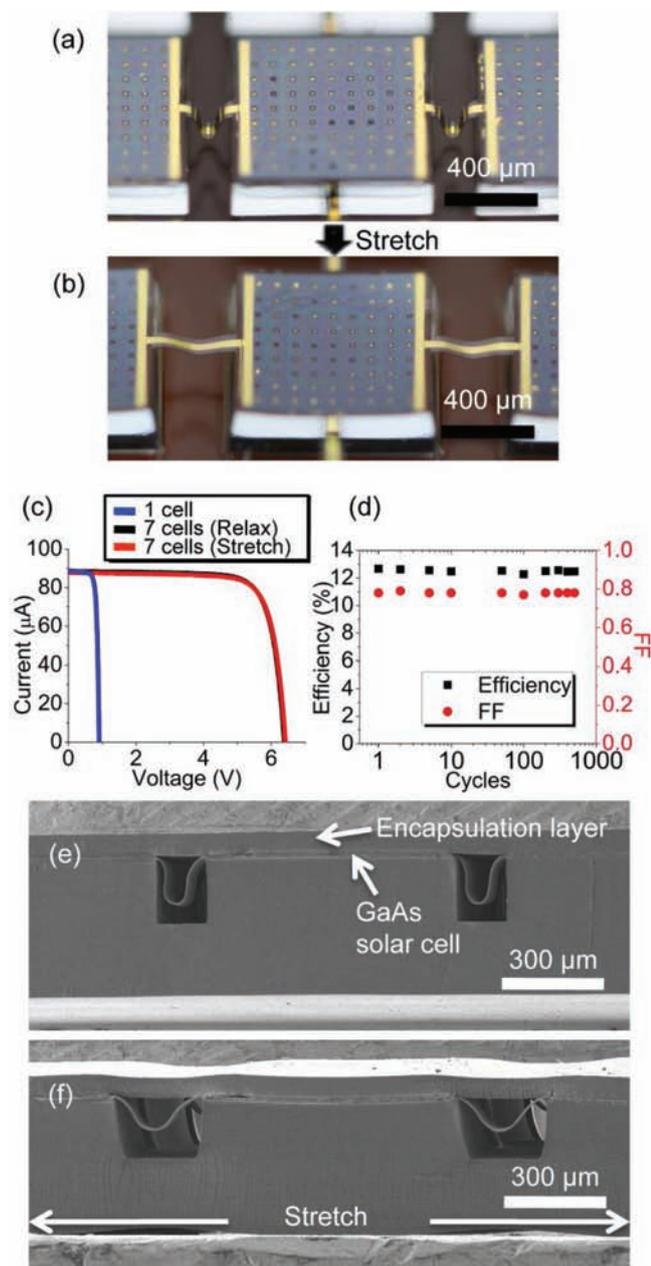


Figure 4. Optical microscope images and electrical characteristics of a representative stretchable GaAs photovoltaic module, and SEM images of encapsulated modules. Optical images of a part of the module in relaxed (a) and stretched (~20% biaxial) (b) states. (c) Current–voltage characteristics of a single microcell in the array (blue) and of seven microcells with series interconnections in relaxed (black) and stretched (red) states. (d) Experimental results of mechanical cycling tests. The data reveal no degradation in performance of the module with more than 500 cycles. (e) SEM cross-sectional image of an encapsulated module. The interconnects are clearly visible as arc-shaped structures in the sealed trenches of the substrate. (f) SEM image of a similar module, stretched uniaxially by 15%. The interconnects move freely to accommodate the applied strain.

and fill factor (FF, ~0.78) of the interconnected microcells are constant over more than 500 cycles. Related devices based on flat substrates offer similarly attractive fatigue response, but they do not allow, simultaneously, similar levels of coverage and stretchability

without leading to high interfacial stresses that can cause adverse effects on the module such as the bowing effect.

Devices designed for realistic use require encapsulating layers to protect the devices and interconnects from the environment. Figure 4e and f presents results of systems encapsulated by thin (~50 μm) layers of PDMS. The procedures (Figure 1) maintain open cavities at the positions of the trenches, to retain the favorable, strain-isolating mechanics and to provide space for unrestricted motion of the interconnects (Figure 4e,f). Detailed FEM results are given in the Supporting Information.

The structural designs reported here are attractive for their simplicity, and their applicability not only to photovoltaics, but also to all other emerging areas of stretchable semiconductor device technologies. Although the structures studied here involve regular, square arrays of islands, other lattice geometries and even irregular configurations retain the same favorable mechanics. Finally, we note that additional refinements of the shapes and layouts for the interconnects as well as those of the islands can lead to further improvements. The additional flexibility in system design should be useful for many applications.

Experimental Section

Integrating the Interconnected Solar Microcells on the PDMS Substrate: A flat piece of PDMS was used to lift the interconnected GaAs solar microcells from their silicon substrate after dissolving the underlying layer of PMMA in acetone. Electron beam evaporation of Ti (5 nm) and SiO₂ (30 nm) on the back surfaces prepared the device for bonding with a prestrained, structured substrate of PDMS. Aligned bonding was accomplished using an assembly of translation and rotation stages, with an optical microscope. Annealing at 100 °C for 10 min enhanced the adhesion. Upon release of the prestrain, most of the interconnections between microcells buckled upward into non-coplanar, arc-shaped bridges. Pressing on the entire system with a flat glass plate caused these bridges to buckle downward into the recessed relief on the PDMS. See Supporting Information for further details on fabrication and measurement.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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