Soft graphene–polymer composite makes an extremely sensitive sensor

uch research has been done on Madding graphene to rigid polymer systems to make them harder or stronger, and to impart electrical conductivity. In contrast, very little has been done to investigate graphene in a soft polymer matrix. Jonathan Coleman, professor of chemical physics at the School of Physics and CRANN at Trinity College Dublin, was intrigued when graduate student Conor Boland proposed adding graphene sheets to Silly Putty-silicone oil cross-linked with boric acid. This soft, viscoelastic material is produced commercially by Crayola as a children's toy. It quickly became clear that graphene-putty or "G-putty" demonstrated remarkable resistivity changes when strained due to a new mechanism based on the breaking of graphene nanosheet contacts and the easy movement of these nanosheets in the soft polymer



Mechanical sensing application of G-putty. Resistance wave forms measured while using G-putty senses the pulse. The inset shows a single period of the pulse wave form, with the characteristic dicrotic notch indicated. Credit: *Science*. material. This study was published in a recent issue of *Science* (doi:10.1126/ science.aag2879).

"Usually you stretch a material and you get a very small change in the electrical properties. The unusual thing about this material is that the change in resistivity is so large," says Coleman, noting that stretching G-putty by 1% can yield a fivefold change in the resistance. "That's a considerable resistance change for a tiny deformation. The sensitivity is really almost unprecedented."

This sensitivity ultimately led the research team to investigate the composite's practical use as a sensor of very small signals, like the pulse in a carotid artery or, remarkably, the light step of a spider (weighing 20 mg) on a G-putty surface. But the main focus of the research was to understand the mechanism that led to this sensitivity.

What they found was that G-putty comprises an isotropic three-dimensional network of millions of graphene nanosheets, with each sheet in contact with three or four others, on average. These contacts make it easy for current to flow from one graphene nanosheet to the next. But, because they are surrounded by a low-viscosity polymer matrix, the connections between graphene sheets are not fixed rigidly in place, so they can be easily broken. "Even the smallest deformation breaks enough connections that you can measure the change in resistance," says Coleman, "and that's what makes the sensor so sensitive."

But the mechanism that provides this sensitivity is not without its problems. Ideally, a sensor should maintain the same level of electrical conductivity or resistivity as long as it remains under the same amount of strain. It can return to its resting resistivity state when the strain is removed. But, because the G-putty polymer matrix is so soft, the graphene sheets begin to move around and reestablish broken contacts while still under strain conditions—an undesirable property for a sensor. A plot of resistivity versus time shows an initial linear increase in resistivity when the initial strain occurs, followed by a gradual decrease as the connections are reestablished.

For now, the researchers can use the linear portion of the graph to measure small, rapid deformations such as caused by the beating pulse in a carotid artery, but the reforming of connections between graphene sheets makes it an ineffective sensor for monitoring longer temporal phenomena. Coleman is philosophical about this incongruity: "To commercialize this sensor, we're going to have to make the interesting property of the material—the extreme mobility of the graphene nanosheets in the polymer matrix—go away, which is a little bit ironic, but that's the way it goes."

Coleman says that the solution to this challenge is known and straightforward, and that his team is already working with G-putty to make it a more versatile sensor for both short and long duration strain conditions. Furthermore, the researchers may already have identified a system that is even more sensitive than G-putty, which will be the subject of future work.

"A scientist's work sometimes requires him to use fantasy and go off the beaten path," says Vincenzo Palermo, a materials scientist at the National Research Council of Italy in Bologna, and leader of the activities on composites of the Graphene Flagship project, who was not involved in this research. "This work targets graphene composites with an unconventional 'toy' material, very well known to adults and kids but not much studied in the composites field. This makes Coleman's excellent results even more interesting for both scientists and [laypersons]."

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