Graphene has electrical and mechanical properties that are promising for electronics and energy storage applications. However, the atomically thick material has a penchant to stick together and crumple up, making it difficult to handle. By attaching graphene sheets to a rubber film, researchers have now found a way to crumple and flatten large areas of the material.

Controlled crumpling and unfolding of graphene could be used to make artificial muscles and stretchable electrodes, and might enable many other applications, says Xuanhe Zhao, a professor of mechanical engineering and materials science at Duke University. Zhao, Markus Buehler at the Massachusetts Institute of Technology, Nicola Pugno at the University of Trento, Italy, and their colleagues reported the advance online in the journal *Nature Materials*.

It is hard to control graphene’s morphology, particularly as researchers make big flakes of the material, says Zhao. He likens the material to wet tissue paper. “One can imagine how difficult it is to crumple and then unfold a piece of thin tissue paper soaked in water,” he says.

The tendency of graphene films to deform can be useful. For instance, wrinkled sheets could be used to make high surface-area electrodes for supercapacitors. Researchers have also found a way to crumple up smooth, microscopic graphene flakes, in the process wrapping them around nanoparticles, creating tiny cargo-carrying sacks that could be used in biomedicine.

However, no one has been able to repeatedly crumple and unfold large pieces of graphene. Zhao and his colleagues wanted to do this because, he says, “such a capability can potentially advance the performance of graphene-based devices and materials, as well as open avenues to exploit new properties of graphene.”
The researchers grew centimeters-wide graphene films on a nickel substrate. Then they used a rubber stamp to transfer it to an acrylic-based sheet that they had pre-stretched to many times its original size. When they let the acrylic relax, the graphene compressed and crumpled, with some parts still attached to the rubber and others detached. Stretching the rubber film unfolded the graphene.

The crumpled graphene sheets remain conductive when the material is repeatedly stretched and twisted, a property that could be useful for making electrodes for actuators and energy-harvesting devices, the researchers say. Such flexible energy harvesters could go in shoes and clothes, converting motion into electricity.

To demonstrate artificial muscles that can expand and contract on demand, the researchers attached graphene to the top and bottom surfaces of a pre-stretched dielectric elastomer. The material doubles in area when the researchers apply a voltage of 3,000V between the graphene films, and returns to its original form when the voltage is removed.

“People usually think of graphene as flat two-dimensional sheets,” says Jiaxing Huang, a materials science and engineering professor at Northwestern University. “This work shows how wrinkly graphene can have new properties unavailable to flat ones.”

Vitor Pereira, a graphene researcher at the National University of Singapore, finds the works interesting because the researchers are able to “neatly manipulate the degree and pattern of the crumpling pattern in two spatial dimensions, and not just along one, which is what has been mostly done so far.” He thinks the material could find use in flexible electronics because it “resists large cycles of straining, relaxing, and twisting with little effect on the electronic transport properties.”

Read the abstract in Nature Materials here.