

features. If massive data sets spanning a wide variety of speech conditions were available, direct synthesis would probably match or outperform a two-stage decoding approach. However, given the data-set limitations that exist in practice, having an intermediate stage of decoding brings information about normal motor function of the vocal-tract articulators into the model, and constrains the possible parameters of the neural-network model that must be evaluated. This approach seems to have enabled the neural networks to achieve higher performance. Ultimately, 'biomimetic' approaches that mirror normal motor function might have a key role in replicating the high-speed, high-accuracy communication typical of natural speech.

The development and adoption of robust metrics that allow meaningful comparisons across studies is a challenge in BCI research, including the nascent field of speech BCIs. For example, a metric such as the error in reconstructing the original spoken audio might have little correspondence to a BCI's functional performance; that is, whether a listener can understand the synthesized speech. To address this problem, Anumanchipalli *et al.* developed easily replicable measures of speech intelligibility for human listeners, taken from the field of speech engineering. The researchers recruited users on the crowdsourcing marketplace Amazon Mechanical Turk, and tasked them with identifying words or sentences from synthesized speech. Unlike the reconstruction error or previously used automated intelligibility measures<sup>6</sup>, this approach directly measures the intelligibility of speech to human listeners without the need for comparison with the original spoken words.

Anumanchipalli and colleagues' results provide a compelling proof of concept for a speech-synthesis BCI, both in terms of the accuracy of audio reconstruction and in the ability of listeners to classify the words and sentences produced. However, many challenges remain on the path to a clinically viable speech BCI. The intelligibility of the reconstructed speech was still much lower than that of natural speech. Whether the BCI can be further improved by collecting larger data sets and continuing to develop the underlying computational approaches remains to be seen. Additional improvements might be obtained by using neural interfaces that record more-localized brain activity than that recorded with electrocorticography. Intracortical micro-electrode arrays, for example, have generally led to higher performance than electrocorticography in other areas of BCI research<sup>3,7</sup>.

Another limitation of all current approaches for speech decoding is the need to train decoders using vocalized speech. Therefore, BCIs based on these approaches could not be directly applied to people who cannot speak. But Anumanchipalli and colleagues showed that speech synthesis was still possible when volunteers mimed speech without making sounds,

although speech decoding was substantially less accurate. Whether individuals who can no longer produce speech-related movements will be able to use speech-synthesis BCIs is a question for future research. Notably, after the development of the first proof-of-concept studies of BCIs to control arm and hand movements in healthy animals, similar questions were raised about the applicability of such BCIs in people with paralysis. Subsequent clinical trials have compellingly demonstrated rapid communication, control of robotic arms and restoration of sensation and movement of paralysed limbs in humans using these BCIs<sup>8,9</sup>.

Given that human speech production cannot be directly studied in animals, the rapid progress in this research area over the past decade — from groundbreaking clinical studies that probed the organization of speech-related brain regions<sup>10</sup> to proof-of-concept speech-synthesis BCIs<sup>6</sup> — is truly remarkable. These achievements are a testament to the power of multidisciplinary collaborative teams that combine neurosurgeons, neurologists, engineers, neuroscientists, clinical staff, linguists and computer scientists. The most recent results would also have been impossible without the emergence of deep-learning and artificial neural networks, which have widespread applications in neuroscience and neuroengineering<sup>11–13</sup>.

Finally, these compelling proof-of-concept demonstrations of speech synthesis in individuals who cannot speak, combined with the rapid progress of BCIs in people with upper-limb paralysis, argue that clinical studies

involving people with speech impairments should be strongly considered. With continued progress, we can hope that individuals with speech impairments will regain the ability to freely speak their minds and reconnect with the world around them. ■

**Chethan Pandarinath and Yahia H. Ali** are in the Wallace H. Coulter Department of Biomedical Engineering, Emory University and Georgia Institute of Technology, Atlanta, Georgia 30322, USA. C.P. is also in the Department of Neurosurgery and at the Emory Neuromodulation and Technology Innovation Center, Emory University. e-mail: chethan@gatech.edu

1. Anumanchipalli, G. K., Chartier, J. & Chang, E. F. *Nature* **568**, 493–498 (2019).
2. Wolpaw, J. R., Birbaumer, N., McFarland, D. J., Pfurtscheller, G. & Vaughan, T. M. *Clin. Neurophysiol.* **113**, 767–791 (2002).
3. Pandarinath, C. *et al.* *eLife* **6**, e18554 (2017).
4. Chartier, J., Anumanchipalli, G. K., Johnson, K. & Chang, E. F. *Neuron* **98**, 1042–1054 (2018).
5. Akbari, H., Khalighinejad, B., Herrero, J. L., Mehta, A. D. & Mesgarani, N. *Sci. Rep.* **9**, 874 (2019).
6. Angrick, M. *et al.* *J. Neural Eng.* <https://doi.org/10.1088/1741-2552/ab0c59> (2019).
7. Gilja, V. *et al.* *Nature Med.* **21**, 1142–1145 (2015).
8. Hochberg, L. R. *et al.* *Nature* **442**, 164–171 (2006).
9. Collinger, J. L., Gaunt, R. A. & Schwartz, A. B. *Curr. Opin. Biomed. Eng.* **8**, 84–92 (2018).
10. Bouchard, K. E., Mesgarani, N., Johnson, K. & Chang, E. F. *Nature* **495**, 327–332 (2013).
11. Yamins, D. L. K. *et al.* *Proc. Natl Acad. Sci. USA* **111**, 8619–8624 (2014).
12. Mathis, A. *et al.* *Nature Neurosci.* **21**, 1281–1289 (2018).
13. Pandarinath, C. *et al.* *Nature Methods* **15**, 805–815 (2018).

## POLYMER CHEMISTRY

## A step towards truly recyclable plastics

**Crosslinked polymer networks known as thermoset plastics have many applications, but can't be reshaped or recycled. A thermoset with reorganizable crosslinks retains its useful properties, but has recyclability built in.**

CORALIE JEHANNO & HARITZ SARDON

The fact that most currently used plastics cannot be easily recycled has produced severe environmental problems, caused considerable losses to the global economy and depleted finite natural resources<sup>1,2</sup>. Of the most widely used plastics today, thermosets are of great interest for high-performance applications, but are particularly impractical for recycling because they cannot be reprocessed using heat or solvents. Writing in *Nature Chemistry*, Christensen *et al.*<sup>3</sup> report thermosets formed using covalent links known as diketoenamines, which can be reorganized within the material's polymer network.

Remarkably, the diketoenamines allow the plastics to be recycled in an energy-efficient process to regenerate pristine monomers, which can then be used to make thermosets that are almost identical to the original material.

Conventional plastics are polymers that are designed to survive ambient onslaughts from light, water, heat and so on. But this resilience also makes them difficult to recycle. Most widely used plastics can be recycled in principle, but reprocessing is costly, energy-demanding and usually produces poor-quality materials. The performance of recycled plastics is therefore typically not as good as that of newly made polymers<sup>4</sup>.

Thermosets are particularly problematic

because the polymer molecules are permanently crosslinked by covalent bonds. This crosslinking means that thermosets have much better solvent resistance and thermomechanical properties than do thermoplastics — a family of polymers that lack molecular crosslinks. Thermosets are therefore highly attractive materials for present and future use in high-temperature electronic devices or in automotive applications, and they currently account for 15–20% of global plastic production<sup>5</sup>. But the fact that they cannot be reshaped or recycled, either by melting or by processing in solution, is a major drawback<sup>6</sup>.

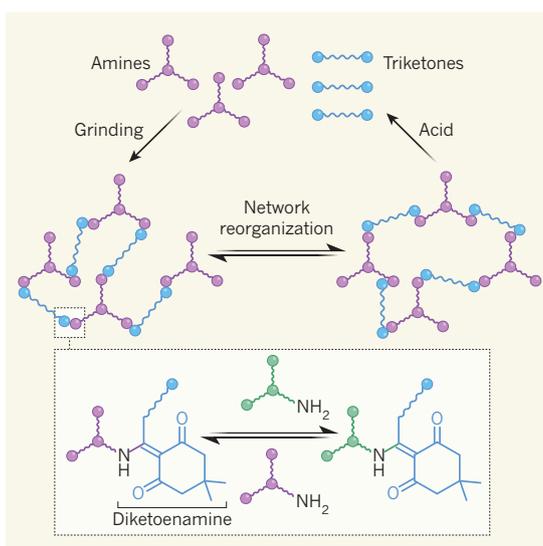
Researchers are therefore trying to make thermosets that can be recycled, reshaped in energy-efficient ways, or both, without compromising the materials' excellent chemical and thermal resistance<sup>7</sup>. One idea is to use reorganizable polymer networks in thermosets<sup>8,9</sup>. Such plastics would have the desirable properties of thermosets under most conditions, but could be reshaped using certain stimuli.

For example, thermosets known as vitrimers have been intensively studied in the past decade. Vitrimers have glass-like fluidity: they flow when heated because the covalent bonds reorganize at high temperatures. This allows vitrimers to be thermally reshaped and eventually recycled, like thermoplastics<sup>10</sup>. But because high temperatures are often required during reprocessing, these dynamic thermosets tend to degrade with each usage cycle, which reduces their utility.

Some thermosets<sup>7</sup> have been designed to depolymerize at low temperatures under acid conditions. This allows the plastics to be used in closed-loop cycles — in which monomers are recovered from the depolymerization and used to remake the plastic<sup>11,12</sup>. Nevertheless, more work is needed to develop polymers that can yield 100% monomer conversion rates, as well as to improve the sustainability of the chemistry and make it suitable for industrial-scale processes, and to find ways of depolymerizing thermosets in mixed plastic waste.

In their search for thermosets to use in closed-loop cycles, Christensen *et al.* designed polymers that form dynamic crosslinked networks based on diketoenamine bonds (Fig. 1). The authors' networks are produced almost spontaneously from starting materials in a solvent-free, versatile process that could, in principle, be used to make many analogous materials. Ketoenamine groups — which differ from diketoenamines by having one carbonyl (C=O) group, rather than two — have previously been shown to reorganize within a polymer network to enable polymer reshaping<sup>13</sup>.

Previous work<sup>14</sup> had shown that ketoenamine-containing polymer networks are stable in acid, but Christensen *et al.* found



**Figure 1 | Dynamic covalent bonds enable monomers to be recovered from a polymer network.** Christensen *et al.*<sup>3</sup> have synthesized a polymer network in which the molecules are crosslinked by diketoenamine bonds. The polymer is made by simply grinding together two monomers (amine and triketone compounds), and it belongs to a useful family of plastics known as thermosets. The crosslinks can reorganize within the polymer because amine (NH<sub>2</sub>) groups from different molecules react with the diketoenamine groups. When the polymer is treated with strong acid, the monomers can be recovered and used to make the polymer again.

that, unexpectedly, their diketoenamine network can be depolymerized by treating it with a strong acid. The process takes less than 12 hours and produces excellent monomer yields, with the difference in reactivity being associated with the presence of the extra C=O group in the diketoenamine crosslinks. The authors found that the monomers could be recovered using an operationally simple procedure, and that they could be reused to make thermosets that had nearly identical properties to the material that had been depolymerized.

Christensen *et al.* went on to carry out the depolymerization of their material in the presence of other widely used plastics and additives (such as fibreglass, colourants or flame-retardants) that are often found in plastic waste that has been collected for recycling. No contamination of the recovered monomers by these materials was observed, demonstrating that the reaction tolerates many different additives. This is a particularly interesting finding because it suggests that the depolymerization method could be used to recycle fibre-reinforced plastics — finding a way to recycle such composites is one of the biggest challenges in the field.

This latest work opens up avenues of research for the preparation of fully recyclable thermosets, but it also raises scientific and technological questions. For example, Christensen and colleagues carried out their polymerizations on a gram scale — can the reactions also be carried out on industrial scales for plastic manufacturing? Moreover, the authors needed

to use large amounts of acidic and basic water to recycle and purify the monomers, so will this process be environmentally friendly on industrial scales?

To facilitate the translation of Christensen and colleagues' strategy from academic experiments to commercial industrial processes, dynamic polymer networks should now be implemented in widely used thermosets such as polyurethane, polyester and epoxide resins. Academics and industrialists will also need to work together to define the costs and benefits of next-generation plastics compared with current polymers, and to carry out life-cycle assessments<sup>15</sup>. This will require chemists to work routinely with other scientists to evaluate the consequences of the entire life cycle of synthesized products.

The development of robust materials that combine excellent chemical and heat resistance with outstanding recycling capabilities could greatly assist the transition from the current linear model of plastics production and consumption — in which limited resources are used to make products that have a finite lifetime and are then disposed of — to a sustainable, circular economy that minimizes waste and maximizes resource use. Next-generation plastics will certainly need to be compatible with closed-loop life cycles to sustainably satisfy the expectations of the world's growing population. In the meantime, work such as this from Christensen *et al.* takes us considerably closer to the synthesis of plastics that have minimal environmental impact. ■

**Coralie Jehanno and Haritz Sardon** are at *POLYMAT, University of the Basque Country UPV/EHU, Joxe Mari Korta Center, 20018 Donostia-San Sebastian, Spain.*  
e-mail: haritz.sardon@ehu.eus

1. Ellen MacArthur Foundation. *New Plastics Economy Global Commitment* (Ellen MacArthur Foundation, 2019).
2. Rahimi, A. & Garcia, J. M. *Nature Rev. Chem.* **1**, 0046 (2017).
3. Christensen, P. R., Scheuermann, A. M., Loeffler, K. E. & Helms, B. A. *Nature Chem.* <https://doi.org/10.1038/s41557-019-0249-2> (2019).
4. Sardon, H. & Dove, A. P. *Science* **360**, 380–381 (2018).
5. Fortman, D. J. *et al. ACS Sustain. Chem. Eng.* **6**, 11145–11159 (2018).
6. Long, T. E. *Science* **344**, 706–707 (2014).
7. Garcia, J. M. *et al. Science* **344**, 732–735 (2014).
8. Montarnal, D., Capelot, M., Tournilhac, F. & Leibler, L. *Science* **334**, 965–968 (2011).
9. Wojtecki, R. J., Meador, M. A. & Rowan, S. J. *Nature Mater.* **10**, 14–27 (2011).
10. Denissen, W., Winne, J. M. & Du Prez, F. E. *Chem. Sci.* **7**, 30–38 (2016).
11. Lloyd, E. M. *et al. Chem. Mater.* **31**, 398–406 (2019).
12. Zhu, J.-B., Watson, E. M., Tang, J. & Chen, E. Y.-X. *Science* **360**, 398–403 (2018).
13. DeBlase, C. R., Silberstein, K. E., Truong, T.-T., Abruna, H. D. & Dichtel, W. R. *J. Am. Chem. Soc.* **135**, 16821–16824 (2013).
14. Kandambeth, S. *et al. J. Am. Chem. Soc.* **134**, 19524–19527 (2012).
15. Rorrer, N. A. *et al. Joule* <https://doi.org/10.1016/j.joule.2019.01.018> (2019).