Comments on 'Enhanced piezoresistivity in Ni-silicone rubber composites'

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This paper describes the accepted meaning of the term 'negative resistance' and its use in the literature, and discusses the features in the previously published data on QTC^{TM} (market name of a metal–polymer composite) that show that the non-linear, hysteretic current voltage characteristics are not caused by Joule heating. The benefit of the high metal filler loading in QTC^{TM} and other unique features of this composite are reviewed.

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Chang Fang-Gao *et al.*[1] in reporting their study of a Ni-silicone conductive composite containing spherical Ni particles have made adverse comments on our research on a composite containing spiky Ni particles (QTC^{TM}) (market name of a metal–polymer composite).[2,3] They criticise our use of the term 'negative resistance' and state that we have used the slope of the current versus voltage plots to calculate sample resistance. Furthermore they assert that the non-linear response of the current to an increasing applied voltage is due to Joule heating in our material. Neither of these comments is valid.

The term 'negative resistance' (or negative differential resistance) is widely used in the literature in a variety of contexts, principally electronic devices.[4–8] It is used to describe a region of the current versus voltage data that has a negative slope but does not imply that the slope is used to determine a resistance value. This was the context in which we used the term. The determination of a resistance is only valid when the current is linearly dependent on the voltage. We state in Ref. [2] that the resistance values quoted were determined in the small current limit, i.e. the linear region of the current versus voltage data. In Ref. [3] values of resistance are quoted for undeformed and highly compressed QTC^{TM} when the current is linearly dependent on the voltage, c.f. Figs. 2 and 3.[3] We do not cite any other values of resistance, but in both papers simply present the non-linear current versus voltage data.

The electrical non-linearity of QTC^{TM} which occurs in other than the undeformed and highly compressed states is not due to Joule heating. We were aware of the possibility that a positive temperature coefficient of resistance could give rise to an increase in sample resistance if the temperature of the sample rose significantly. In Ref. [2] we comment that this has been observed to result in a small region of current decreasing as voltage increases in a carbon black filled composite.[9] The high metal loading of the QTC^{TM} samples means that they have good thermal conductivity. The samples were compressed between two large metal blocks, which acted as heat sinks and limited the temperature rise in the samples. In Ref. [3] we note that the highly compressed sample subjected to 2.5 A at 0.5 V shows a small shift in resistance due to Joule heating; this effect is so small that it is barely discernable in the current versus voltage data.

Examination of the current versus voltage plots shows that the non-linearity and hysteresis cannot be due to Joule heating. As the voltage increases the current increases super-linearly up to a point close to the maximum power dissipation, a sub-linear increase would occur if Joule heating is significant. In the region where the current falls as the voltage increases, the current shows large, rapid fluctuations. These can be a large fraction of the flowing current and occur on timescales shorter than one second. These are too large and fast to be due to fluctuations in sample temperature. The levels of power dissipation on the for-

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ward and reverse voltage sweeps are similar and can even be larger during the reverse sweep. Figure 8 of Ref. [2] shows maximum power dissipation as the voltage increases of about 8 W while on the reverse sweep it is larger than 8 W. Despite this, on the up sweep the current falls as the voltage increases beyond the point of maximum dissipation while on the reverse sweep the current remains high as the voltage decreases. Since Joule heating must give rise to an increase in the effective resistance of the sample it cannot be the origin of the non-linear, hysteretic behaviour observed for QTC^{TM}.

Chang *et al.*[1] make comparisons between their composite and QTC^{TM}. These include a lower Ni loading and higher strain sensitivity as determined by calculated gauge factors. The materials and method of fabrication used by Chang *et al.* will result in a conventional percolative composite. When the composition is close to the percolation threshold, compression will result in a large decrease in sample resistance as the sample traverses the percolation transition. This effect was reported recently by Abyaneh and Kulkarni[10] who studied composites containing spherical Ni particles with a range of metal loadings. This was also the situation in the materials studied by Martin *et al.*[11] Unfortunately, close to the percolation threshold the sample resistance and the response to pressure become sensitive to the metal loading.[12] QTC^{TM} is loaded well above the percolation threshold and in consequence the variation of resistance with deformation is much less sensitive to small variations in composition. Because of the high loading, QTC^{TM} also displays large changes in resistance when subjected to elongation, bending and twisting.[2] We do not know of any other composite with similar properties.

In comparing calculated gauge factors Chang *et al.*[1] have quoted a value for their composite in the region of greatest sensitivity at a compressive strain of 0.78%. The gauge factor for QTC^{TM} is calculated from our data taken at much larger deformations. These correspond to a region where the change in resistance as a function of deformation for Chang *et al*'s sample 2702# is much lower than at 0.78% strain. At deformations comparable to those we reported[2] the gauge factor of sample 2702# would also be lower than the value quoted. This fact is not made clear in their paper.

Finally we note that the principal use of the form of QTC^{TM} described in our publications[2,3] is as a switch in wearable electronics and similar applications. This is possible because QTC^{TM} can be compressed to give resistances in the milliohm range, much smaller than those achieved by Chang *et al.* Despite the high compression involved,[3] QTC^{TM} is not damaged and rapidly recovers its initial high resistance. The QTC^{TM} has been subjected to one million switching cycles without any degradation in performance, demonstrating durability under these extreme conditions. Durability and reproducibility are crucial factors in any commercial application; these aspects are not touched on by Chang *et al.*

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