Comment on "Two-step PTC effect in immiscible polymer blends filled with carbon black" by Weihua Di, Guo Zhang, Yi Peng and Zhudi Zhao, *Journal of Materials Science*, 39, 695–697, 2004

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Frydman first observed the positive temperature coefficient (PTC) effect in a carbon black (CB)-filled low-density polyethylene composite in 1945 [1]. After that, many studies were performed on CB-filled semicrystaline composite due to its potential application in industry such as overcurrent protection in circuit [2–9]. It is well accepted by the scientific community that the strong PTC effect of CB-filled semicrystalline polymer composites is caused by an increase in the average interparticle or aggregates distance of CB due to the large thermal expansion as a result of the melting of polymer crystallites [7]. It was also found that when the temperature is above the T_m of the polymers, a sharp decrease in resistivity occurs, which is termed the negative temperature coefficient (NTC) effect. It has been suggested that the NTC effect of a CB-filled single semicrystalline polymer composite is caused by the formation of a flocculated structure when the viscosity of the polymer is sufficiently low at elevated temperatures.

It should be strongly stressed that most past investigations on the PTC and NTC effects of CB-filled polymer composites were focused on the composites containing CB and a single semicrystalline polymer. Few studies were conducted on the PTC and NTC effects of CB-filled immiscible semicrystalline polymer composites [10–14]. In our previous studies of CB-filled PP/UHMWPE and CB-filled ETFE/HDPE composites, two resistivity jumps were observed, and designated as the double PTC effects. The mechanism of the double PTC effect was also studied. For example, for CB-filled ETFE/HDPE composite, the large thermal expansion owing to the melting of HDPE and ETFE crystallites must be responsible for the first and second PTC effects, respectively [12].

Recently, a paper entitled "Two-step PTC effect in immiscible polymer blends filled with carbon black" has been published in *Journal of Materials Science* (39 (2004), 695–697). It has been found that this paper is very similar to my paper [12] in many aspects, namely: in the technical idea, the preparation procedure of CB-filled immiscible polymer blends, the proposed mechanism of double PTC effects, and even in similar sentence construction in many places.

First, it has been found that the technical idea in the paper is almost the same as that in our paper. The only difference between their idea and our idea is that they just replaced high melting point semicrystalline polymer ETFE by another high melting semicrystalline polymer PP. Even the CB concentration is the same. As a consequence, Fig. 3 in their paper [15] is very similar to Fig. 1 in my paper [12], and Fig. 4 in their paper [15] is almost the same as Fig. 1 in my paper, except that the temperatures corresponding the two resistivity jumps are different.

Second, it also has been observed that the preparation procedure of CB-filled immiscible polymer blends is almost the same as that in our paper. In the second paragraph in experimental section of my paper [12], it is stated:

The CB-filled ETFE/HDPE composite was prepared by melt-mixing the materials in a Haak mixer at 280 °C and 30 rpm for 15 min. The compound obtained was further compressed into 2-mm sheet by a hot press at 280 °C and 16 MPa.

In the fourth paragraph in their paper [15], the following sentences appear:

The CB-filled and a mixture of CB and carbon fiber (CF)-filled HDPE/PP composites were prepared by melt-mixing the materials in a Brabender mixer at 180 °C and 30 rpm for 10 min. The compound obtained was further compressed into 1.5-mm sheet in a vulcanizing press at 180 °C.

Third, for the mechanism for 10 wt% N660-filled ETFE/HDPE composite, we have proposed that the melting of HDPE and ETFE crystallites and the two sharp resistivity jumps occur at similar temperatures. Therefore, the large volume expansion owing to the melting of HDPE and ETFE crystallites must be responsible for the first and second PTC effects, respectively [12]. In their paper, for 10 wt% CB-filled 40/60 HDPE/PP blends, they suggested that the melting of HDPE and PP crystallites and the two sharp resistivity jumps occur at the similar temperatures. Therefore, the large volume expansion owing to the melting of HDPE and PP crystallites is responsible for the two-step PTC effect [15]. Accordingly, it is very clear that their mechanism is totally the same as ours not only in physics but also in the manner in which it was expressed.

As shown above, even though there are many similarities between their paper [15] and mine [12], the authors of the paper [15] did not mention my paper [12]. The reasons for this are unclear.

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Apart from those mentioned above, in another my paper [11], in the eighth paragraph in the Results and Discussion section, it is stated:

When the temperature increases to the T_m of UHMWPE, the large volume expansion of UHMWPE due to the melting of the UHMWPE crystallites breaks up most of the type I+M conductive pathways, resulting in a major jump in the resistivity. As the temperature further increases to the T_m of PP, a small jump in the resistivity occurs. This increase in the resistivity can be attributed to the volume expansion of the PP phase as a result of the melting of the PP crystallites.

In the eighth paragraph in their paper [15], appear the following sentences:

When the temperature increases to the T_m of PE, the large volume expansion of PE due to the melting of crystallites breaks down the conductive paths in the PE phase, resulting in the happening of the first resistivity jump. As the temperature further increases to the T_m of PP, the second resistivity jump occurs, which is attributed to the volume expansion of PP, breaking down the conductive paths at the PE/PP interface.

Obviously, the logic structure is the same, and the construction of the sentences is nearly the same.

In summary, the paper [15] is very similar to my papers [11, 12]. The similarities are in many aspects including technical idea, preparation procedure of CB-filled immiscible polymer blends, the proposed mechanism of the double PTC effects, and even English writing. This is unacceptable.

References

1. E. FRYDMAN, UK Patent Spec. 604 659 I718 14S (1945).
2. F. KOHLER, US Patent 3 243 753 (1966).
3. K. OHE and Y. NATIO, *Jpn. J. Appl. Phys.* **10** (1971) 199.
4. J. MEYER, *Polym. Engng. Sci.* **13** (1973) 462.
5. G. KLASON and J. KUBAT, *J. Appl. Polym. Sci.* **19** (1978) 831.
6. M. NARKIS, A. RAM and F. FLASHNER, *Polym. Engng. Sci.* **18** (1978) 649.
7. A. VOET, *Rubb. Chem. Techn.* **54** (1981) 42.
8. R. D. SHERMAN, L. M. MIDDLEMAN and S. M. JACBOS, *Polym. Engng. Sci.* **23** (1983) 36.
9. M. AL-ALLAK, A. W. BRINKMAN and J. WOODS, *J. Mater. Sci.* **28** (1993) 117.
10. J. Y. FENG and C. M. CHAN, *Polym. Engng. Sci.* **39** (1999) 1207.
11. *Idem.*, *Polymer.* **41** (2000) 4559.
12. *Idem.*, *ibid.* **41** (2000) 7279.
13. I. MIRONI-HARPAZ and M. NARKIS, *J. Polym. Sci. Part B.: Polym. Phys.* **39** (2001) 1415.
14. *Idem.*, *Polym. Engng. Sci.* **41** (2001) 205.
15. W. DI, G. ZHANG, Y. PENG and Z. D. ZHAO, *J. Mater. Sci.* **39** (2004) 695.

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