

# CARBON FIBER POLYMER-MATRIX COMPOSITE INTERFACES AS THERMOCOUPLE JUNCTIONS

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## Introduction

A thermocouple is a thermoelectric thermometric device that involves a junction between two dissimilar materials. The voltage between the two dissimilar materials at the ends away from the junction relates to the temperature difference between the junction and these ends. The physics lies on the Seebeck effect, i.e., the movement of the mobile charge carriers from the hot point to the cold point of each dissimilar material and the consequent voltage difference between the hot and cold points of each dissimilar material.

The dissimilar materials used for thermocouples are conventionally metals. This paper provides thermocouples in the form of continuous carbon fiber polymer-matrix composites. Carbon fibers are used because of their electrical and thermal conductivity. The interlaminar interface (i.e., interface between adjacent laminae in a composite) serves as the junction in the thermocouple. The dissimilar materials are laminae with different types of carbon fibers (fibers that differ in carrier type and/or concentration).

The advantages of fiber composite thermocouples compared to conventional thermocouples are low cost, mechanical ruggedness, processability into various shapes and sizes, and that the thermocouple is itself the structure. The last advantage means that the structure is itself thermocouples, thus making the structure be able to monitor its temperature without the need for embedded or attached devices. This translates to low cost, high durability, large sensing volume and absence of mechanical property degradation (which occurs in the case of embedded sensors).

The Seebeck effect involving a single type of material rather than dissimilar materials has been reported in carbon fibers (no matrix) [1-3] and in carbon fiber composites [4,5]. The use of dissimilar materials allows the voltage measurement to be made only at one end (say the end at room temperature) of the dissimilar materials, thus making thermocouples convenient to be used. Furthermore, appropriate selection of the dissimilar materials can make the change in measured voltage per unit rise in temperature

(i.e., thermocouple sensitivity) larger than the Seebeck coefficient of a single type of material.

Carbon fibers can be n-type or p-type even without intercalation. Intercalation greatly increases the carrier concentration, thus making the fibers strongly n-type or strongly p-type, depending on whether the intercalate is an electron donor or an electron acceptor. Although there had been study of the thermopower of intercalated carbon fibers [3], n-type and p-type forms of carbon fibers have not been exploited as dissimilar materials for thermocouples. This work shows that such thermocouples are particularly sensitive.

One of the drawbacks of intercalated graphite is the instability over time, either due to intercalate desorption or reaction with environmental species. For the case of bromine (acceptor) as the intercalate, the instability due to desorption can be overcome by the use of a residue compound, i.e., a compound that has undergone desorption as much as possible so that the remaining intercalate is strongly held, thereby making the compound stable. The stability of bromine intercalated carbon fibers has been previously demonstrated [6-8]. For the case of an alkali metal such as sodium (donor) as the intercalate, the instability due to reactivity with moisture can be overcome by the use of an alkali metal hydroxide (with the alkali metal ions in excess) as the intercalate [9]. Therefore, this paper uses bromine as the acceptor intercalate and sodium hydroxide (with  $\text{Na}^+$  ions in excess) as the donor intercalate.

Although considerable attention has been given to intercalated carbon fibers, little attention has been given to composites that involve these fibers [10-12]. Previous work on these composites has been focused on the electrical conductivity, due to the relevance to electromagnetic interference shielding and other applications. In contrast to previous work, this paper addresses the thermoelectric behavior of the composites, particularly composites involving dissimilar carbon fibers that meet at a junction to form a thermocouple.

## Experimental methods

The carbon fibers used were Thornel P-25, P-100 and P-120 2K pitch-based fibers (Amoco Performance Products, Alpharetta, GA) and T-300 PAN-based fibers (in the form of 976 epoxy unidirectional fiber prepregs, Hy-E 1076E, ICI Fiberite, Tempe, AZ).

Intercalation was carried out only for P-100 and P-120 fibers, due to their relatively high crystallinity. Bromine intercalation involved exposure to bromine vapor in air at room temperature for 10 days, followed by desorption in a fume hood at room temperature for several months. Sodium hydroxide intercalation involved immersion of the fibers in a liquid solution of NaOH and molten sodium contained in a nickel crucible.

Thermocouple junctions were epoxy-matrix composite interlaminar interfaces. In this study, a junction was formed by allowing two laminae to overlap partially and then curing the stack under heat and pressure, as required for the curing of the epoxy matrix. The overlap region served as the junction; the remaining regions served as thermocouple wires.

## Results and discussion

Pristine P-25 is slightly n-type, pristine T-300 is strongly n-type. A junction comprising pristine P-25 and pristine T-300 has a positive thermocouple sensitivity that is close to the difference of the Seebeck coefficients (or the absolute thermoelectric powers) of T-300 and P-25, whether the junction is unidirectional or crossply. Pristine P-100 and pristine P-120 are both slightly n-type. Intercalation with sodium causes P-100 and P-120 to become strongly n-type. Intercalation with bromine causes P-100 and P-120 to become strongly p-type. A junction comprising bromine intercalated P-100 and sodium intercalated P-100 has a positive thermocouple sensitivity that is close to the sum of the magnitudes of the absolute thermoelectric powers of the bromine intercalated P-100 and the sodium intercalated P-100. Similarly, a junction comprising bromine intercalated P-120 and sodium intercalated P-120 has a positive thermocouple sensitivity that is close to the sum of the magnitudes of the absolute thermoelectric powers of the bromine intercalated P-120 and the sodium intercalated P-120.

A junction comprising n-type and p-type partners has a thermocouple sensitivity that is close to the sum of the magnitudes of the absolute thermoelectric powers of the two partners. This is because the electrons in the n-type partner as well as the holes in the p-type partner move away from the hot point toward the corresponding cold point. As a result, the overall effect on the voltage difference between the two cold ends is additive. By using junctions comprising strongly n-type and strongly p-type

partners, a thermocouple sensitivity as high as  $+82 \mu\text{V}/^\circ\text{C}$  was attained.

## Conclusion

Thermocouples made from n-type carbon fibers (e.g., sodium intercalated P-100 fibers) and p-type carbon fibers (e.g., bromine intercalated P-100 fibers) in the form of epoxy-matrix composites, using the interlaminar interface as the thermocouple junction, were found to exhibit thermocouple sensitivity up to  $82 \mu\text{V}/^\circ\text{C}$  – close to the sum of the magnitudes of the Seebeck coefficients of the two partners of the thermocouple. Bromine intercalation changed the Seebeck coefficient (with copper as the reference) of P-100 fibers from  $+1.7$  to  $-43 \mu\text{V}/^\circ\text{C}$ . Sodium intercalation changed it from  $+1.7$  to  $+48 \mu\text{V}/^\circ\text{C}$ . Similarly large effects were observed for intercalated P-120 fibers. Pristine fibers gave similar junctions, but with a much smaller value of the thermocouple sensitivity. The thermocouple sensitivity was the same for unidirectional and crossply junctions.

## References

- [1] di Vittorio SL, Dresselhaus MS, Endo M, Issi JP, Piraux L, Bayot V. *J Mater Res* 1991; 6(4):778-783.
- [2] Kuriyama K, Dresselhaus MS. *J Mater Res* 1991; 6(5):1040-1047.
- [3] Tsukamoto J, Takahashi A, Tani J, Ishiguro T. *Carbon* 1989;27(6):919-923.
- [4] Stokes KL., Tritt TM, Fuller-Mora WW, Ehrlich AC, Jacobsen RL. *Int. Conf. Thermoelectrics, ICT, Proc.* 1996;164-167.
- [5] Sun M, Li Z, Mao Q, Shen D. *Cem Concr Res* 1998;28(4):549-554.
- [6] Ho CT, Chung DDL. *Carbon* 1990;8(6):825-830.
- [7] Gupta V, Mathur RB, Bahl OP, Marchand A, Flandrois S. *Carbon* 1995;33(11):1633-1639.
- [8] Wessbecher DE, Forsman WC, Gaier JR. *Synth Met* 1988;26(2):185-194.
- [9] Hérolde C, Hérolde A, Lagrange P. *J Phys Chem Solids* 1996;57(6-8):655-662.
- [10] Gaier JR, Hamburger PD, Slabe ME, *Carbon* 1991;29(3):313-320.
- [11] Gaier JR, Davidson ML, Shively RK, *Technology Transfer in a Global Community, Int. SAMPE Tech. Conf., vol. 28, 1996, SAMPE, Covina, CA, p. 1136-1147.*
- [12] Katsumata M, Endo M, Yamanashi H, Ushijima H. *J Mater Res* 1994;9(7):1829-1833.
- [13] Pollock DD. *Thermoelectricity: Theory, Thermometry, Tool, ASTM Special Technical Publication 852, Philadelphia, PA, 1985, p. 121.*