Flexible graphite as a heating element

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Abstract

Flexible graphite is an effective heating element. It provides temperatures up to 980 °C (though burn-off occurs in air at 980 °C), response half-time down to 4 s, and heat output at 60 s up to 5600 J. The electrical energy for heating by 1 °C is 1–2 J in the initial portion of rapid temperature rise. The temperature and heat output increase with decreasing thickness and with increasing power.
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1. Introduction

Flexible graphite is a flexible sheet made by compressing a collection of exfoliated graphite flakes (called worms) without a binder [1]. During exfoliation, an intercalated graphite (graphite compound with foreign species called the intercalate between some of the graphite layers) flake expands typically by over 100 times along the *c* axis. Compression of the resulting worms (like accordions) causes the worms to be mechanically interlocked to one another, so that a sheet is formed without a binder.

Due to the exfoliation, flexible graphite has a relatively large specific surface area (e.g. 15 m^2/g [2]). As a result, flexible graphite is used as an adsorption substrate [3,4]. Due to the absence of a binder, flexible graphite is essentially entirely graphite, other than the ash and the residual amount of decomposed intercalate (such as sulfur in the case of sulfuric acid being the intercalate). As a result, flexible graphite is chemically and thermally resistant, and low in coefficient of thermal expansion (CTE). Due to its microstructure involving graphite layers that are preferentially parallel to the surface of the sheet, flexible graphite is high in electrical and thermal conductivities in the plane of the sheet [5,6]. Due to the graphite layers being somewhat connected perpendicular to the sheet (i.e. the honeycomb microstructure of exfoliated graphite), flexible graphite is electrically and thermally conductive in

the direction perpendicular to the sheet (although not as conductive as the plane of the sheet) [5,6]. These in-plane and out-of-plane microstructures result in resilience and impermeability to fluids perpendicular to the sheet. The combination of resilience, impermeability and chemical and thermal resistance makes flexible graphite attractive for use as a gasket material for high temperature or chemically harsh environments.

Gasketing (i.e. packaging, sealing) [7–11] is by far the main application of flexible graphite, which can replace asbestos. Other than gasketing, a number of applications have emerged recently, including adsorption [3,4], electromagnetic interference (EMI) shielding [2], vibration damping [12], electrochemical applications [13] and stress sensing [14].

Graphite has long been used as a heating element. In addition to graphite in a monolithic form [15], pyrolytic graphite deposited on boron nitride has been used [16]. Furthermore, polymer–matrix composites containing carbon fibers [17] or carbon black [18], and carbon–matrix composites [19] have been used. Flexibility or shape conformability of the heating element is desirable for many applications, such as the deicing of aircraft [20,21] and the heating of floors, pipes and boilers. Flexible graphite is thus attractive. It is also attractive because it is in a sheet form, is corrosion-resistant, does not need to be encased and does not need machining for shaping. In contrast, conventional graphite requires expensive machining to attain the shape required for the heating element. This paper evaluates the effectiveness of flexible graphite as a heating element. Although the use of flexible graphite as a

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heating element has been mentioned in patents [22,23], evaluation of the effectiveness has not been reported.

Previous work on the electrical resistivity of flexible graphite is limited to the resistivity at room temperature or below [24–27]. The resistivity decreases with increasing temperature in the temperature range from about 10 to 300 K [24], but it increases with increasing temperature in the temperature range from 0.1 to 6 K [25]. The low temperature behavior is relevant to fundamental study of the conduction mechanism. However, for the resistance heating application, the resistivity above room temperature is relevant.

2. Experimental

Flexible graphite sheets (Grade GTB) of thickness ranging from 0.13 to 1.17 mm were provided by EGC Enterprises, (Mentor, OH, USA). The specific surface area is 15 m^2/g, as determined by nitrogen adsorption using the Micromeritics (Norcross, GA, USA) ASAP 2010 instrument. Based on geometric consideration, this specific surface area corresponds to a crystallite layer height of 0.18 μm within a sheet. According to the manufacturer, the ash content of flexible graphite is <5.0%; the density is 1.1 g/cm^3; the tensile strength in the plane of the sheet is 5.2 MPa; the compressive strength (10% reduction) perpendicular to the sheet is 3.9 MPa; the thermal conductivity at 1093 °C is 43 W/m K in the plane of the sheet and 3 W/m K perpendicular to the sheet; the coefficient of thermal expansion (CTE) (21–1093 °C) is -0.4×10^{-6}/°C in the plane of the sheet.

DC electrical resistivity measurements were conducted in the in-plane direction of flexible graphite using the four-probe method. A Keithley 2001 multimeter was used. Samples were in the form of rectangular bars of size 100×5 mm and thickness ranging from 0.13 to 1.17 mm. Each electrical contact was applied around the entire perimeter of the bar. The four contacts were at four parallel cross-sectional planes perpendicular to the length of the bar. The outer two contacts (for passing current) were 80 mm apart; the inner two contacts (for measuring the voltage) were 60 mm apart. The resistivity was measured at and above room temperature. Heating was achieved by using a resistance heater and a temperature controller, which provided a heating rate of 5 °C/min.

Evaluation of flexible graphite as a heating element was conducted by passing a fixed DC current (ranging from 2 to 10 A) along the length of the specimen (100×5 mm in size and thickness ranging from 0.13 to 1.17 mm) by using electrical contacts (80 mm apart) in the form of silver paint in conjunction with copper wire. The voltage drop (ranging from 0.2 to 10 V) along the length of the specimen was measured by using two other electrical contacts (60 mm apart), also in the form of silver paint in conjunction with copper wire. During the test, a weight was applied to the top surface of the specimen in order to provide electrical contacts in the form of pressure contacts, since the silver paint degraded and lost its adhesive ability as the specimen became hot. The weight was electrically insulated from the specimen by using zirconia fiber cloth. The temperature of the specimen was measured as a function of time during constant current application and in the subsequent period in which the current was off by using a K-type thermocouple located in the middle of the top surface of the specimen. The constant current period was long enough for the temperature to essentially level off to a maximum.

3. Results and discussion

3.1. Electrical resistivity

Fig. 1 shows the variation with temperature of the in-plane electrical resistivity of flexible graphite of two different thicknesses. The resistivity decreases with increasing temperature for any thickness, as expected [24]. It does not vary systematically with the thickness for a given temperature. The Arrhenius plot of the logarithm of the conductivity versus the inverse of the absolute temperature is linear (Fig. 2) and its slope gives an activation energy of electrical conduction of 0.01 eV. This low value is consistent with the crystallinity of the flexible graphite material [28].

3.2. Heating element evaluation

Fig. 3 shows the change in temperature with electrical energy, which is proportional to time, since the power (product of voltage and current) is constant, for two power levels and a thickness of 0.13 mm. For a given power, the
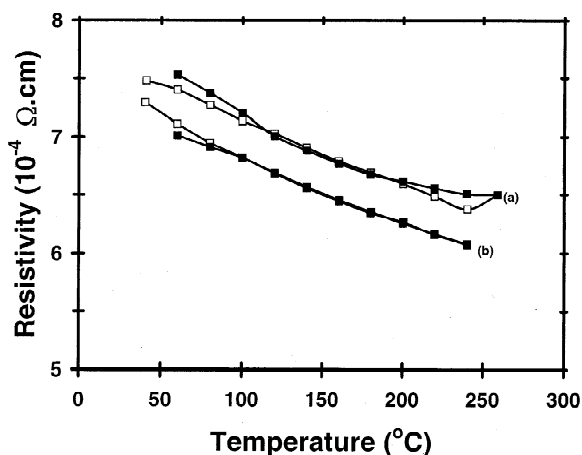

Fig. 1. In-plane volume electrical resistivity vs. temperature during heating (open squares) and cooling (solid squares) for thicknesses (a) 0.13 mm and (b) 0.38 mm.
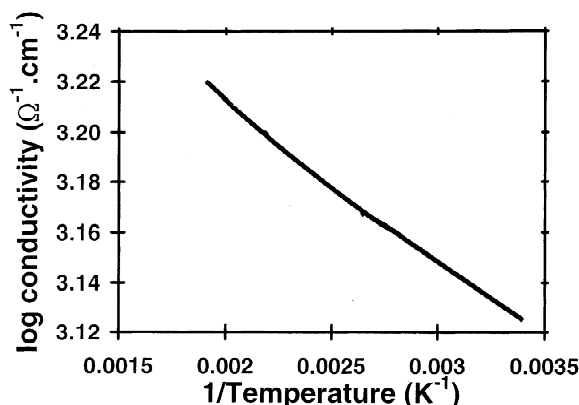
Fig. 2. Arrhenius plot of log conductivity vs. inverse absolute temperature during heating for thickness 0.38 mm.
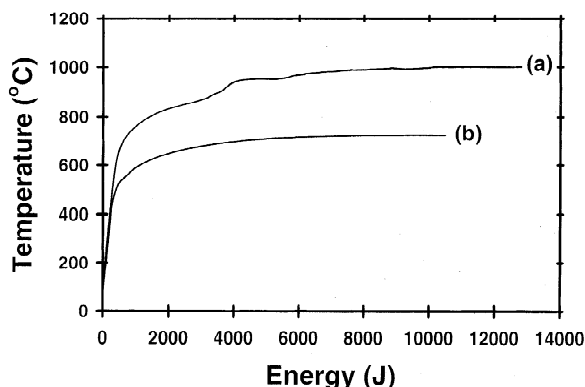

Fig. 3. Temperature vs. electrical energy during heating for thicknesses 0.13 mm and power (a) 94.1 W (9.68 V, 9.72 A) and (b) 52.2 W (6.74 V, 7.75 A).

temperature increases with energy, as expected, but it levels off gradually. For the same energy, a higher power gives a higher temperature. The data in Fig. 3, together with those for other combinations of power and thickness (Table 1), show that the energy needed per °C temperature rise in the initial portion (10–15 s) of rapid temperature rise is quite independent of the thickness and power.

Fig. 4 shows the change in temperature with time in the constant current period and in the subsequent period in which the current is off for two power levels and a thickness of 0.13 mm. The heating is faster than the cooling. The maximum temperature increases with decreasing thickness (Table 1), due to the higher resistance associated with a lower thickness. It increases with increas-

Table 1
Performance of flexible graphite as a heating element

Thickness (mm)	Power (W)	Maximum temperature (°C)	Time to reach half of maximum temperature (s)	Time to cool to half of maximum temperature (s)	Energy to heat by 1 °C[a] (J)	Heat output at 60 s (J)
0.13	4.07	108	6	4	1.04	239
0.13	14.6	282	5	6	1.08	861
0.13	31.8	518	5	7	1.14	1880
0.13	52.2	711	4	7	1.15	3091
0.13	94.1	981	4	10	1.25	5588
0.38	1.43	53	6	8	1.43	80
0.38	6.39	149	8	8	1.52	360
0.38	11.74	229	6	8	1.24	666
0.38	21.3	354	5	9	1.38	1210
0.38	31.2	492	6	17	1.86	1785
0.51	0.93	45	6	7	2.10	53
0.51	4.12	89	8	10	1.90	234
0.51	9.56	171	12	8	2.29	545
0.51	16.75	252	9	11	2.16	957
0.51	23.76	327	7	9	2.32	1360
0.76	0.96	33	7	7	2.22	53
0.76	4.07	82	13	8	1.48	224
0.76	8.37	144	5	9	2.25	459
0.76	15.2	219	6	11	1.75	842
0.76	22.6	314	7	10	2.18	1258
1.17	0.48	36	15	11	1.03	24
1.17	1.78	61	9	10	0.87	87
1.17	4.39	109	11	12	1.19	222
1.17	6.88	148	11	15	1.21	352
1.17	10.62	193	9	15	1.55	551

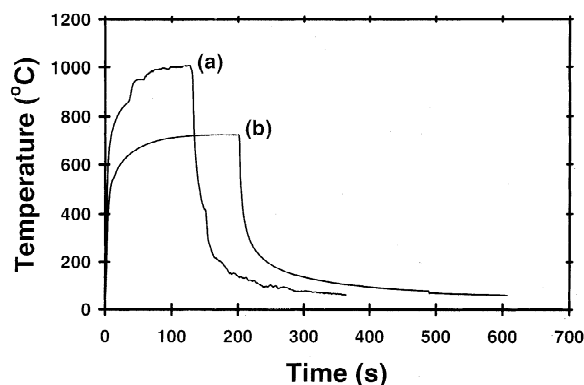[a] In the initial portion (10–15 s) of rapid temperature rise.

Fig. 4. Temperature vs. time during heating and subsequent cooling for thickness 0.13 mm and power (a) 94.1 W (9.68 V, 9.72 A) and (b) 52.2 W (6.74 V, 7.75 A).

Fig. 6. Heat output vs. time in the initial portion for thickness 0.38 mm and power (a) 31.2 W (3.13 V, 9.97 A), (b) 11.7 W (2.03 V, 5.78 A).

ing power (Table 1 and Fig. 4), as expected. The response time during heating (time to reach half of the maximum temperature) tends to decrease slightly with decreasing thickness; it does not vary monotonically with the power (Table 1). The response time during cooling (time to cool to half of the maximum temperature) increases with increasing power for the same thickness, except for the thickness of 0.51 mm, at which no clear trend was observed (Table 1).

The highest temperature attained is 981 °C, as obtained by using the smallest thickness and the highest power. The flexible graphite glows in this case. The temperature increases associated with the fluctuations in the curve for this thickness in Figs. 3(a) and 4(a) are attributed to burn-off resulting from oxidation of the graphite [29,30]. The burn-off leads to an increase in resistance and thus a rise in temperature. Because of the burn-off, it is not

desirable to use flexible graphite as a heating element in air at a power as high as 94 W. At a lower power of 52 W (Fig. 3(b) and 4(b)), the fluctuations are absent in the curves, due to the relatively minor extent of burn-off. The use of flexible graphite as a heating element in an inert atmosphere will remove the burn-off problem.

The heat output is given by the electrical energy input minus the heat absorbed by the heating element (i.e. flexible graphite). The heat absorbed is given by the product of the specific heat, mass and temperature change. Assuming that the specific heat is constant at 830 J/kg K (the value for graphite [31]), the heat output was calculated, as shown in Figs. 5 and 6 for different combinations of thickness and power. After the initial portion (5 s, Fig. 6), the heat output increases linearly with time, since the electrical energy input also increases linearly with time for a given power. For the same thickness and time, the heat output increases with increasing power, since the electrical energy input increases with increasing power. For essentially the same power and the same time, the heat output essentially does not vary with the thickness, as shown in Table 1 for a time of 60 s.
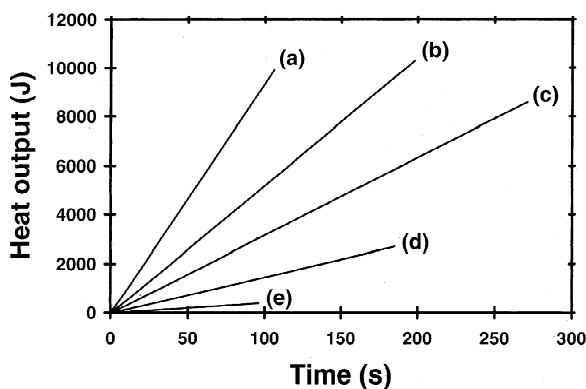

Fig. 5. Heat output vs. time for thickness 0.13 mm and power (a) 94.1 W (9.68 V, 9.72 A), (b) 52.2 W (6.74 V, 7.75 A), (c) 31.8 W (5.29 V, 6.01 A), (d) 14.6 W (3.79 V, 3.86 A), and (e) 4.07 W (2.13 V, 1.91 A).
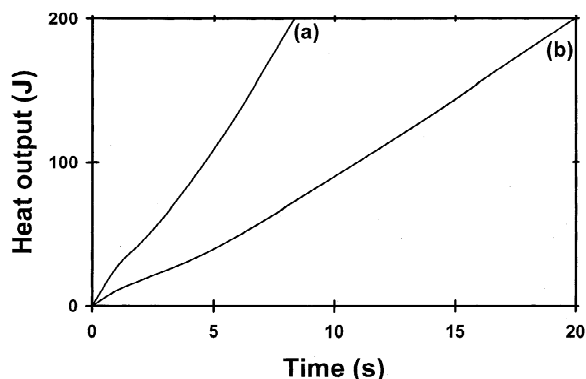
4. Conclusion

Flexible graphite is effective as a heating element, as shown by passing a DC current in the in-plane direction. It provides temperatures up to 980 °C (though burn-off occurs in air at 980 °C), with a response half-time as low as 4 s. The heat output at 60 s of heating is up to 5600 J, and the electrical energy to heat by 1 °C in the initial portion of rapid temperature rise is 1–2 J. Both the temperature and the heat output increase with decreasing thickness (0.13–1.17 mm) and with increasing power (0.48–94.1 W).

Acknowledgements

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