THE SEEBECK COEFFICIENT OF GRAPHITE OR THE STB MODEL REVISITED

C. A. Klein and R. N. Donadio
Raytheon Research Division, Waltham, Massachusetts

For the purpose of investigating the thermoelectric power of graphite we consider it a fair approximation to describe the majority-carrier bands by means of

$$E_{e,h} = E_0(\xi) \pm \hbar^2\kappa^2/2m^*(\xi), \quad (1)$$

where $E_0(\xi) = 2\gamma_2\cos^2(\xi/2)$ and $m^*(\xi) = (4/3)(\hbar/a_0)^2\,(\gamma_1/\gamma_0^2)\cos(\xi/2)$. In terms of this model, and using standard notations, the free carrier concentration amounts to

$$n_e + n_h = \frac{\langle m^*\rangle}{2\pi\hbar^2 c_0}\int_{-\pi}^{+\pi}\cos(\xi/2)d\xi \int_{-\infty}^{+\infty}|E - E_0(\xi)|(-\frac{\partial f_0}{\partial E})dE\,, \quad (2)$$

which is found to be almost independent of γ_2 for $kT \geqslant \gamma_2$. Since we are concerned with temperatures in the range above 300°K, we conclude that for all practical purposes we may set $E_0(\xi) \equiv 0$, or in other words, ignore the effects of "overlap" in the frame of this work.

With $a = n_h/n_e$ and $b = \mu_e/\mu_h$, the Seebeck coefficient of graphite (layer-plane configuration) is best expressed as follows:

$$\alpha_{\parallel} = (a\alpha_h + b\alpha_e)/(a + b), \quad (3)$$

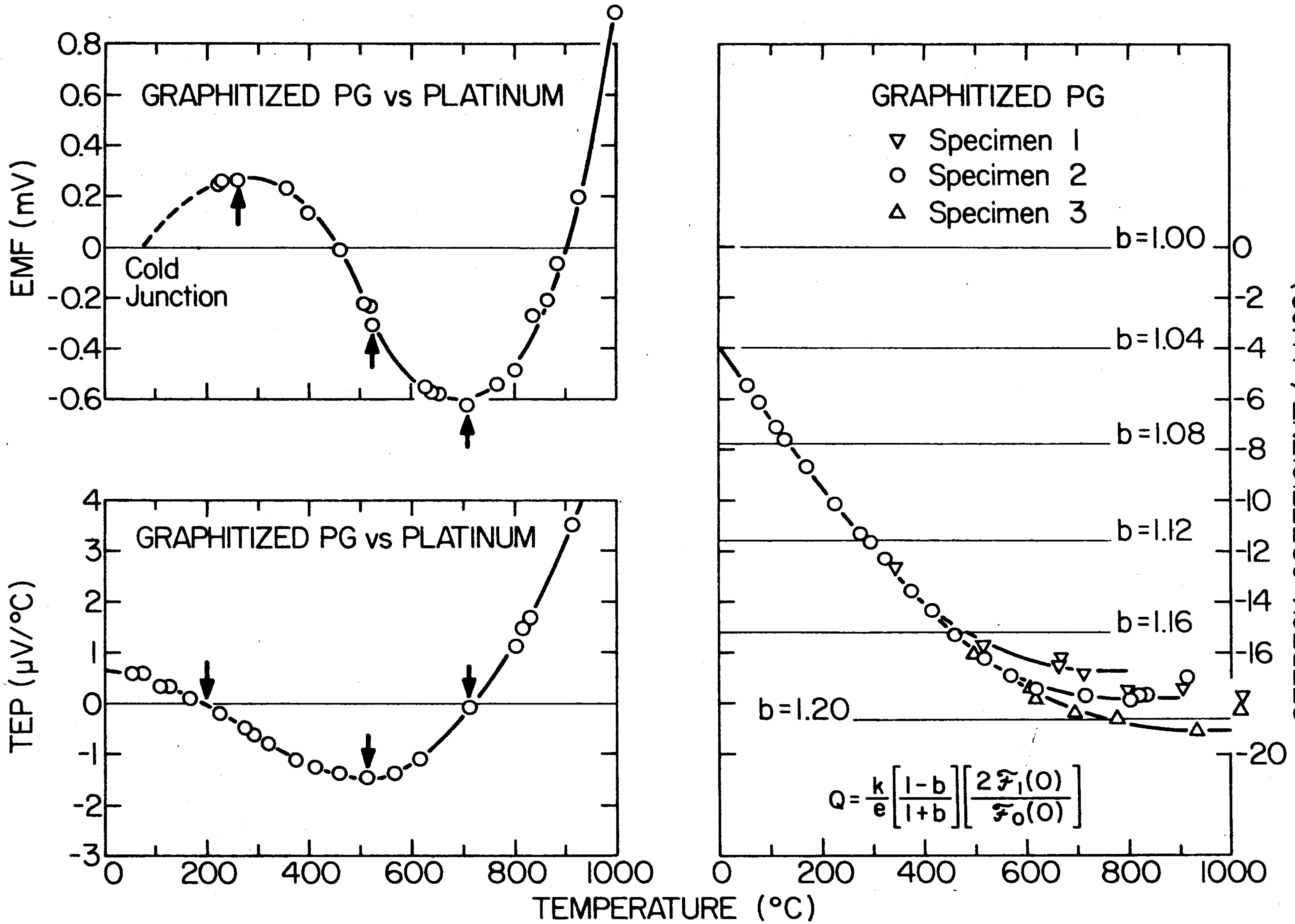
where $\alpha_e = -\frac{k}{e}(\frac{K_1}{kTK_0} - \frac{E_F}{kT})$ and $K_i = \frac{1}{4\pi^3}\int_{BZ}\tau_e v_x^2\, E^i(-\frac{\partial f_0}{\partial E})d^3k$ —– similar expressions hold for the hole contribution α_h. Following McClure [Proc. Fifth Carbon Conf. 2, 3(1963)], we assume that the relaxation times may be derived from the "golden rule" for transition probabilities and thus that, within the approximation $\gamma_2 \equiv 0$, we have energy-independent relaxation times. On this basis it is a straightforward matter to demonstrate that

$$\alpha_{\parallel} = (k/e)(a + b)^{-1}\,[\,a(\delta_+ - \Delta/kT) - b(\delta_- + \Delta/kT)\,] \quad (4)$$

with $\delta_\pm = [\,2\mathcal{F}_1(\pm\Delta/kT)\,]\,[\mathcal{F}_0\,(\pm\Delta/kT)\,]^{-1}$, where $\mathcal{F}_j$ designates a Fermi-Dirac integral and Δ measures the Fermi-level depression ($\Delta = -E_F$).*

We will show that data obtained in the range 0° to 1000°C for various classes of pyrolytic graphite (PG) substantiate Eq.(4) and lead to the following conclusions: (a) In graphitized PG the mobility ratio b increases slowly with temperature and may reach 1.2 at 800°C (see attached figure); (b) In turbostratic PG the Seebeck coefficient behaves in accordance with a Fermi level depressed by ≈ 0.015 eV and changes sign above 1000°C; (c) In boronated PG the temperature dependence of $\alpha_{\parallel}$ differs drastically with boron content though metallic characteristics emerge at B/C ⩾ 0.3 %.

* This accords with a previous result of Klein [J. Appl. Phys. 35, 2947 (1964)], but does no longer involve unrealistic assumptions such as spherical energy surfaces and acoustic lattice scattering.



GRAPHITIZED PG vs PLATINUM
EMF (mV)
Cold
Junction
GRAPHITIZED PG vs PLATINUM
TEP (μV/°C)
GRAPHITIZED PG
Specimen 1
Specimen 2
Specimen 3
b=1.00
b=1.04
b=1.08
b=1.12
b=1.16
b=1.20
SEEBECK COEFFICIENT (μV/°C)
$Q = \frac{k}{e}\left[\frac{1-b}{1+b}\right]\left[\frac{2\mathcal{F}_1(0)}{\mathcal{F}_0(0)}\right]$
TEMPERATURE (°C)