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## Electrical contact resistance in filaments

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Electrical contact resistance (ECR) influences the electrochemical performance of porous electrodes made of stacked discrete materials (e.g., carbon nanotubes, nanofibers, etc.) for use in supercapacitors and rechargeable batteries. This study establishes a simple elasticity-conductivity model for the ECR of filaments in adhesive contact. The elastic deformation and size of electrical contact zone of the filaments are determined by using an adhesive contact model of filaments, and the ECR of adhesive filaments is obtained in explicit form. Dependencies of the ECR upon the filament geometries, surface energy, and elasticity are examined. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4718019>]

Thin fibers and filaments typically carry high strength, large specific surface area, and excellent manufacturability superior to their bulk counterparts. Fibrous materials have found broad applications in thermal/sound insulators, gas/fluid filters, catalyst carriers, tissue scaffolds, and various paper products. With the recent intensive research in energy conversion and storage technologies, conductive fibrous materials, e.g., carbon nanotubes (CNTs), carbon micro/nanofibers, nanowires/nanorods of metal oxides, and conductive polymer fibers, have been considered as excellent candidates as porous electrodes for use in supercapacitors and rechargeable batteries due to their large specific surface area (for more charge storage and faster charge/discharge rate) and favorable electrical connectivity.<sup>1–8</sup> For instance, Fig. 1 shows a typical multi-walled CNT network under consideration for use in porous electrodes of supercapacitors. To function as charge collector as well as current-delivery channel in electrochemical energy conversion and storage devices, fibrous materials are expected to be reliable in both electrical conductivity and structural integrity. To date, a variety of investigations have been devoted to understanding the mechanical behavior of planar and spatial fiber networks since the pioneering works by van Wyk<sup>9</sup> and Cox.<sup>10</sup> Several follow-ups<sup>11–19</sup> have also been made to exploration of the effective stiffness of fiber networks in different extent of elaboration. In addition, for ultrathin compliant fibers (e.g., polymer nanofibers produced by electrospinning), surface effect may also influence their mechanical response such as the effective axial tensile modulus,<sup>20</sup> dynamics,<sup>21,22</sup> adhesive contact,<sup>23,24</sup> surface instability,<sup>25</sup> and wetting properties,<sup>26–29</sup> among others.

When fibrous materials are integrated into electrodes for use in supercapacitors and rechargeable batteries, their electrical connectivity and conductivity directly influence the effective electrical, thermal, and electrochemical performances of the electrodes.<sup>30</sup> Recently, a few studies have been initiated on measurements and modeling of the contact conductivity of metallic foams and fiber networks used in

fuel cells, heat exchangers, etc.<sup>31–34</sup> In parallel, Barber<sup>35</sup> formulated an elastic/electrical analogy between mechanical and electrical contacts, by which the electrical contact resistance (ECR) between elastic bodies in contact can be determined. In addition, Kogut and Komvopoulos<sup>36,37</sup> calculated the ECR between two fractal surfaces via solving the consecutive contact mechanics and electrostatic field problems numerically. Yet, no study has been reported so far on the ECR of filaments in contact, especially ultrathin nanofibers and CNTs. In principle, the ECR between ultrathin filaments depends upon the type of contact, electrical conductivity and geometries of the filaments. In this work, a simple electrical contact model is to be formulated for ultrathin filaments in adhesive contact. The filaments are assumed to be isotropic and linearly elastic. A three-dimensional (3D) adhesive contact model of filaments<sup>23</sup> is adopted for determining the contact properties, in which the Derjaguin-Muller-Toporov (DMT) assumption<sup>38</sup> is used and the adhesive force is estimated via Bradley's approach;<sup>39,40</sup> the elastic deformation and size of adhesive contact zone are calculated within the framework of Hertz contact theory.<sup>41–43</sup> Barber's elastic/electrical analogy<sup>35</sup> is further to bridge the ECR and adhesive contact of the filaments.

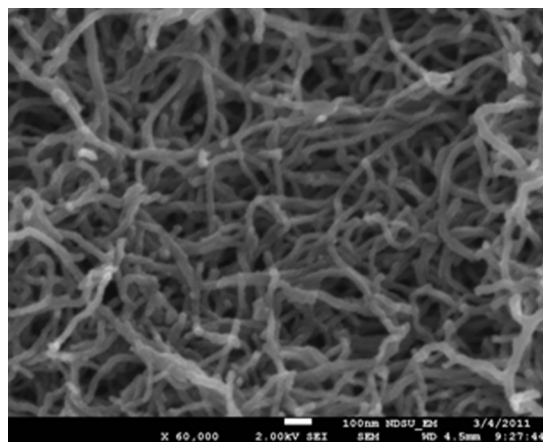


FIG. 1. Carbon nanotube assembly as porous electrode for use in supercapacitors

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Given two conductive elastic bodies in contact, the contact area is a function with respect to the material properties, surface asperities, and contact force exerted between the bodies. The corresponding ECR varies with the change of contact area due to the magnitude of the contact force. According to the elastic/electrical contact analogy, Barber formulated a relationship for the ECR  $R_c$  via the incremental mechanical contact stiffness:<sup>35</sup>

$$R_c = M(\rho_1 + \rho_2)/(dF/d\Delta), \quad (1)$$

where  $\rho_1$  and  $\rho_2$  are the resistivities of the two bodies in contact, respectively,  $F$  is the external compressive force,  $\Delta$  is the compressive deformation, and  $M$  is the composite modulus of the contacting pair defined by

$$1/M = (1 - \nu_1)/G_1 + (1 - \nu_2)/G_2. \quad (2)$$

In the above,  $G_i$  and  $\nu_i$  ( $i = 1, 2$ ) are the shear moduli and Poisson's ratios of the two bodies in contact, respectively. As illustrated in Fig. 2(a), two ultrathin smooth filaments in contact due to the adhesive force are considered.<sup>39,40</sup> By selecting a proper  $(x, y)$ -coordinate system of the contact zone (tangential to the two surfaces in contact) with respect to the normal  $z$ -axis, the distance between two points at the two curvilinear surfaces out of the contact zone can be approximated as [Fig. 2(b)]

$$z_1 + z_2 = Ax^2 + By^2, \quad (3)$$

where  $A$  and  $B$  are coefficients relating the principal curvatures and the angle between the planes of principal curvatures of the two surfaces in contact. In the case of two identical circular cylindrical filaments of radius  $R$ ,  $A$  and  $B$  can be determined as

$$A = (1 - \cos\varphi)/(2R), \quad B = (1 + \cos\varphi)/(2R), \quad (4)$$

where  $\varphi$  is the spatial angle between the filament axes. Relation (4) indicates that  $A$  and  $B$  are positive, thus, all points with the same mutual distance  $z_1 + z_2$  will be located on an ellipse. Thus, the contact zone of two filaments is an ellipse.

The adhesive force between ultrathin filaments in contact can be estimated according to Bradley's approach<sup>23,39,40</sup> by treating the adhesive force between two unit areas as a long-range Lennard-Jones force:

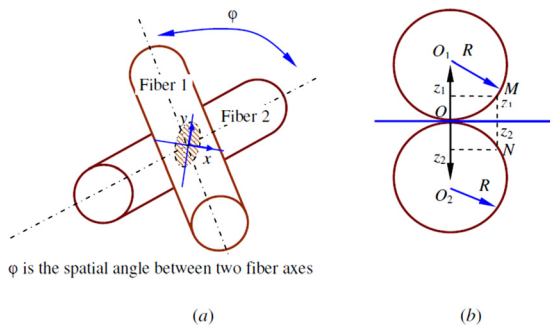


FIG. 2. Two circular cylindrical filaments in contact: (a) elliptical contact zone between filaments ( $x$ - and  $y$ -coordinate corresponding to the two principal axes of the elliptical contact zone) and (b) coordinate systems in cross-section.

$$\sigma(z) = \frac{8\Delta\gamma}{3\varepsilon} \left[ \left( \frac{\varepsilon}{z} \right)^3 - \left( \frac{\varepsilon}{z} \right)^9 \right]. \quad (5)$$

In the above,  $\varepsilon$  is a phenomenological distance between two particles (e.g., atoms or molecules);  $z$  is the distance between two unit areas; and  $\Delta\gamma$  is Dupré adhesion energy defined by

$$\Delta\gamma = \gamma_1 + \gamma_2 - \gamma_{12}, \quad (6)$$

where  $\gamma_1$  and  $\gamma_2$  are the surface energies of the filaments, respectively, and  $\gamma_{12}$  is the interface energy. In the case of two identical filaments in contact,  $\Delta\gamma = 2\gamma$ . Thus, the adhesive force can be estimated via integration of Eq. (5) with respect to the area out of the contact zone (i.e.,  $Ax^2 + By^2 \geq \alpha$  with  $\sqrt{\alpha/A}$  and  $\sqrt{\alpha/B}$  as the semi-axes of the contact ellipse to be determined)<sup>23</sup>

$$F = \frac{8\Delta\gamma}{3\varepsilon} \int_{Ax^2 + By^2 \geq \alpha} \left[ \left( \frac{\varepsilon}{Ax^2 + By^2 - \alpha + h_0} \right)^3 - \left( \frac{\varepsilon}{Ax^2 + By^2 - \alpha + h_0} \right)^9 \right] dA, \quad (7)$$

where  $h_0$  is the minimum gap between surfaces in contact which can be chosen as  $h_0 = \varepsilon$  with the assumption of Bradley's approach. Substitution of Eq. (4) into Eq. (7) yields the adhesive force between two identical, smooth, circular cylindrical filaments in contact as

$$F = 4\pi\gamma R/\sin\varphi. \quad (8)$$

The relation (8) is identical to the adhesive force between two rigid circular cylinders of identical radius, i.e., the adhesive force independent of the size of contact zone. Thus, elastic deformation does not alter the adhesive force between filaments within Bradley's approach.

With the adhesive force [Eq. (8)], the shortening  $\Delta$  between two axes of identical filaments in contact can be determined by Hertz contact theory<sup>23,43</sup> as

$$\Delta = \frac{6\pi}{\sin\varphi} \frac{\gamma R}{G/(1-\nu)} \int_0^\infty \frac{d\xi}{\sqrt{(a^2 + \xi^2)(b^2 + \xi^2)}}. \quad (9)$$

In the above, the semi-axes  $a$  and  $b$  of the elliptical contact zone can be determined as<sup>23</sup>

$$a = m \sqrt[3]{\frac{3\pi(1-\nu)R^2\gamma}{\sin\varphi G}}, \quad b = n \sqrt[3]{\frac{3\pi(1-\nu)R^2\gamma}{\sin\varphi G}}, \quad (10)$$

where the dimensionless coefficients  $m$  and  $n$  depend only upon the filament orientation angle  $\varphi$  (Ref. 42). For instance,<sup>42</sup> for  $\varphi = 0^\circ$ ,  $m = \infty$ ,  $n = 0$ ; for  $\varphi = 45^\circ$ ,  $m = 1.926$ ,  $n = 0.604$ ; for  $\varphi = 90^\circ$ ,  $m = 1.0$ ,  $n = 1.0$ . With the aid of relation (10), the electrical contact area between the filaments is

$$A_e = \pi ab = mn \left[ \frac{3\pi(1-\nu)R^2\gamma}{\sin\varphi G} \right]^{2/3}. \quad (11)$$

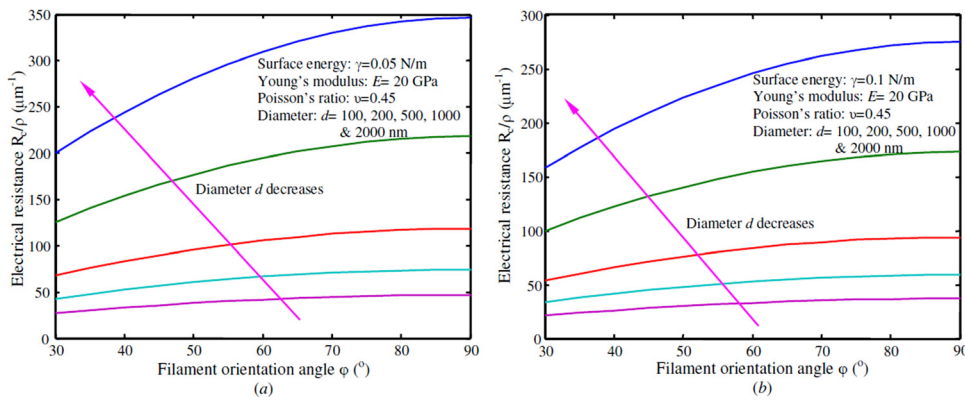


FIG. 3. Variation of the normalized electrical contact resistance  $R_c/\rho$  vs. the orientation angle  $\phi$  for filaments with the surface energies of (a)  $\gamma = 0.05$  N/m and (b)  $\gamma = 0.1$  N/m and the diameters of 100 nm, 200 nm, 500 nm, 1  $\mu$ m, and 2  $\mu$ m, respectively.

Consequently, substituting Eqs. (8) and (9) into Eq. (11) and letting  $G_1 = G_2 = G$ ,  $v_1 = v_2 = v$ , and  $\rho_1 = \rho_2 = \rho$  for identical filaments yield the ECR:

$$R_c = \frac{\rho}{2} \int_0^\infty \left[ 3 - \left( \frac{a^2}{a^2 + \xi^2} + \frac{b^2}{b^2 + \xi^2} \right) \right] \frac{d\xi}{\sqrt{(a^2 + \xi^2)(b^2 + \xi^2)}}. \quad (12)$$

The above relation indicates that the ECR between two adhesive filaments is correlated to the filament resistivity  $\rho$  and the area of adhesive contact zone which is a function with respect to the filament radius  $R$ , orientation angle  $\phi$ , elastic properties  $E$  and  $v$ , and surface energy  $\gamma$  according to Eq. (10). Hereafter, we examine these dependencies by performing numerical experiments. To do so, two filament surface energies  $\gamma = 0.05$  N/m and 0.1 N/m are considered, respectively; for linearly elastic deformation, the Young's modulus  $E$  and Poisson's ratio  $v$  of the filaments are selected as  $E = 20$  GPa and  $v = 0.45$ , thus, the shear modulus is  $G = E/[2(1 + v)] = 6.897$  GPa. These values are close to those of typical polymer fibers. In the calculation, the filament diameters are selected as  $d = 2R = 100$  nm, 200 nm, 500 nm, 1000 nm, and 2000 nm, respectively. Variations of the normalized ECR  $R_0$ , defined by  $R_0 = R_c/\rho$  (unit:  $\mu\text{m}^{-1}$ ), with respect to the orientation angle  $\phi$  between two filaments are plotted in Fig. 3.

From Fig. 3, it can be observed that given the material properties (i.e.,  $E$ ,  $v$ , and  $\gamma$ ) and orientation angle  $\phi$  of the filaments, the ECR of a pair of filaments in adhesive contact increases with decreasing filament diameter. In contrast, at a fixed filament diameter, the ECR increases nonlinearly with increasing  $\phi$ . This tendency is due to the fact that the contact area decreases with increasing  $\phi$  as evidenced by Eq. (11). At small filament diameters, such an increase tendency becomes more obvious since the adhesive contact becomes more obvious at small fiber diameters. Furthermore, Fig. 3 also demonstrates that the surface energy  $\gamma$  plays a noticeable rule in the ECR, especially for filaments at the small diameters. The larger the surface energy  $\gamma$ , the lower the ECR is. This observation is due to the fact that given the material properties (i.e.,  $E$  and  $v$ ) and geometries (i.e.,  $R$  and  $\phi$ ) of the filaments, a larger surface energy corresponds to a higher adhesive force that yields a larger adhesive contact area as indicated in Eq. (11), i.e., a lower ECR. In addition, the present model can be extended conveniently for ECR of other types

of adhesive contact. Yet, to date, no experimental data have been reported in the literature for comparison with the present ECR model although this model has given qualitatively reasonable predictions of the ECR for adhesive filaments. Without a doubt, ECR experiments are expected and of critical importance to developing efficient and reliable porous electrodes for use in modern electrochemical energy conversion and storage devices.

In summary, a simple ECR model of adhesive filaments has been formulated, which gave a rational explanation of the ECR of ultrathin filaments in adhesive contact. The quantitative description of the present model could be used to guide the scaling analysis, measurements, and effective suppression of ECR of porous electrodes made up with stacked discrete particles and filaments. In addition, due to the small size of the filaments, the effect of surface asperity on the ECR is not taken into account. In experiment, such effect is difficult to be quantified, especially in the case of nano-sized filaments. Consequently, the present study also expects innovative ECR experiments that can be used to quantify the ECR of filaments and other elastic bodies in adhesive contact.

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