



Macroscopic networks of thermal conduction: Failure tolerance and switching processes

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ARTICLE INFO

Article history:

Received 17 September 2017

Received in revised form 27 November 2017

Accepted 4 January 2018

ABSTRACT

The world is so wonderful and colorful thanks to the analytic discontinuities and singularities of phase transitions. Here we propose a class of macroscopic networks of thermal conduction that combine regular networks with macroscopic thermally conductive systems. We find that their phase transition phenomena (variation tendency) cannot be explained simultaneously by existing theories of networks, percolation or effective media. We report the *bond-free property* of these networks and the associated three switching processes caused by the geometric property of bonds, and we reveal the effect of single-point connection. Also, we propose some potential applications including thermal diodes. Our results are confirmed by finite-element method simulation and experiment. This work offers different insights into the theories of networks, percolation and effective media. It also provides a different method to design thermal metamaterials and manipulate thermal conductivities.

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1. Introduction

Since the models of small-world networks and scale-free networks [1,2] were proposed at the end of the last century, much breakthrough has been made in the field of network science. As a young discipline, its research contents can be combined with physics, computer science, biology and sociology. However, heat conduction is a traditional field, which can be described by Fourier's law in macroscopic situations. It is interesting to ask what people may get if these two fields are combined. In fact, one can use the method of heat conduction to develop the network theory, as what recommendation model does [3,4], or can directly study the characteristics of heat conduction in regular or complex networks [5–15]. Based on the latter idea, a lot of effort have been paid to explain the abnormal phenomenon in microscopic heat transport, such as thermal rectification, flux localization and interfacial resistance [5–11]. However, studies concerning macroscopic thermal networks [12–15] are not as many as the microscopic ones, and most of them are focused on particulate and related multiphase systems [13–15].

Here we come up with a different network model obeying Fourier's law $\mathbf{J} = -\kappa \nabla T$ (where \mathbf{J} is heat flux density, κ is thermal

conductivity, and ∇T is temperature gradient) and study its percolation (variation tendency), known as an important phenomenon in testing the structural robustness of networks [16–20]. Different from the conventional method which only concerns valid/invalid information of the nodes and/or bonds, we conduct our research via finite-element method (FEM) simulation, which calculates 2-dimensional temperature distribution for each node/bond and contains much more detailed information. Due to the information advantage of FEM, it provides more research results that cannot be obtained from the conventional method. We focus on the connectedness and node influence of the whole network under different conditions of bonds, and point out the role of single-point connections in regular thermally conductive networks. As expected, we find that with different connectivities, our thermally conductive networks show different tolerance to node failures. Furthermore, variation tendencies against node failures under different bond conditions [see Figs. 2(a), 3(a) and 4(a)] can be explained by the theory of network (the network will be more stable with larger connection degrees), percolation [it's consistent with the critical points referred in Fig. 3(c)] and effective medium (continuous change), respectively. The simulation results in Figs. 2–4 help to reveal three switching processes due to the lengths and widths of the bonds. These switching processes are reasonable reasons for tiny changes of the geometry properties of the bonds can cause huge distinctions shown in Figs. 2–4. In other words, the switching processes serve as connections between these three theories.

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As it is crucial to develop efficient methods to manipulate heat flux freely at the background of energy crisis, it seems promising to design thermal metamaterials [21] and manipulate thermal conductivities on the basis of the thermally conductive network. We also propose two potential applications at the end of the manuscript as prototypes to achieve these aims.

2. Simulations and results

2.1. The definition of effective thermal conductivity (κ_{eff}) of thermally conductive networks

In order to study the properties of thermally conductive networks, we need firstly to define their effective thermal conductivity κ_{eff} properly. Taking 4-degree connected network as an example, we illustrate two methods to define the κ_{eff} of such networks. The first method is a series connection method. As illustrated in Fig. 1(a), κ_{eff} of the network can be calculated as

$$\kappa_{\text{eff}} = \kappa_{\text{ref}} \frac{T_{\text{hot}} - T_{\text{mid}}}{T_{\text{mid}} - T_{\text{cold}}} \quad (1)$$

The second method is a heat flux method. As illustrated in Fig. 1(b), the effective thermal conductivity of the network can be obtained by solving the Fourier's law

$$\kappa_{\text{eff}} = J \frac{\Delta x}{T_{\text{hot}} - T_{\text{cold}}} \quad (2)$$

Theoretically, we will get exactly the same κ_{eff} by these two methods. However, there exists slight difference. For the heat flux method, there might be certain deviation when COMSOL is estimating the heat flux J around tiny spaces. So, the first method is more accurate and reliable. We get all the relevant results using this method.

2.2. Results of three thermally conductive networks

First, we consider networks with 3,4,5,6-degree connectivity shown in Fig. 2(a), in which the triangles, squares, pentagons and hexagons stand for thermally conductive nodes while the rectangles connecting neighboring nodes stand for thermally conductive bonds. All the bonds are set as 0.2 cm in length and 0.05 cm in width, and the edge lengths of regular polygon nodes in 3,4,6-degree connected networks are set as 0.2 cm. However, regular pentagon cannot tile a plane according to the tessellation theory. So, in order to consist with Fig. 3(a), for the 5-degree connected network, edge lengths of the nodes are set as 0.2, 0.2, 0.2, 0.2 and 0.1464 cm in order to tile the plane. We use the commercial software COMSOL (<https://www.comsol.com/>) livelink for MATLAB (<https://www.mathworks.com/>) to simulate the thermally conductive properties of these structures. The thermal conductivity (denoted as κ) of each node is set as 0.026 W/(m·K) (which is the κ of air and can be regarded as failure nodes) or 400 W/(m·K) (which is the κ of red copper and can be regarded as normal nodes). Meanwhile, the thermal conductivities of bonds are all set as 4000 W/(m·K) (which is approximately the κ of graphene and is large enough comparing to that of the nodes). Total size of the 3,4,6-degree connected networks is 50×50 crystal cells, and the size of the 5-degree connected network is 20×20 crystal cells (each crystal cell contains 8 nodes and 20 bonds, and 20×20 crystal cells contains more than 10,000 geometries. Concerning there will be a lot of repeated calculation, it's already a pretty large computing burden). We have verified by simulation that the absolute size of the network-structured domain does not affect the effective thermal conductivity of the entire structure. All the boundaries are set as thermal insulation except for the interfaces and hot/cold sources.

For each simulation, we stochastically choose certain percentage of nodes as failure nodes [0.026 W/(m·K)] and others normal

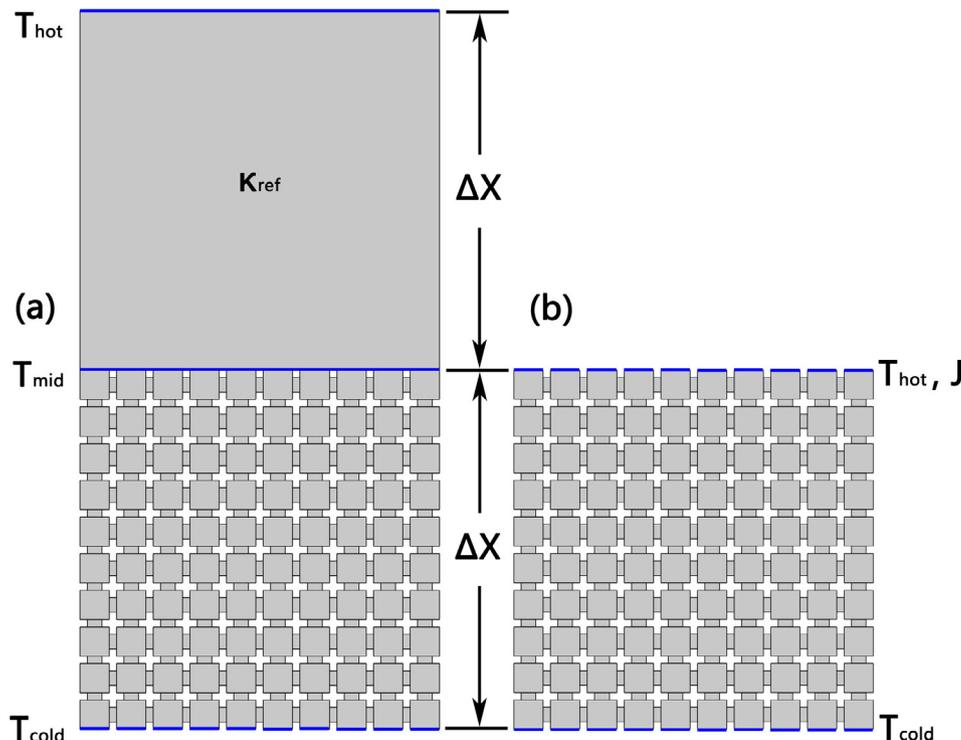


Fig. 1. Two methods to define the effective thermal conductivity. (a) Series connection method. The network is connected with a referential material with the same size and a known thermal conductivity κ_{ref} . The upper boundary of the referential material is set as hot source. The lower boundaries of the network are set as cold sources. The average temperature on the middle line T_{mid} can be obtained by the FEM simulation. (b) Heat flux method. Upper and lower boundaries of the network are set as hot and cold sources. Heat flux on the upper boundary can be obtained by the FEM simulation.

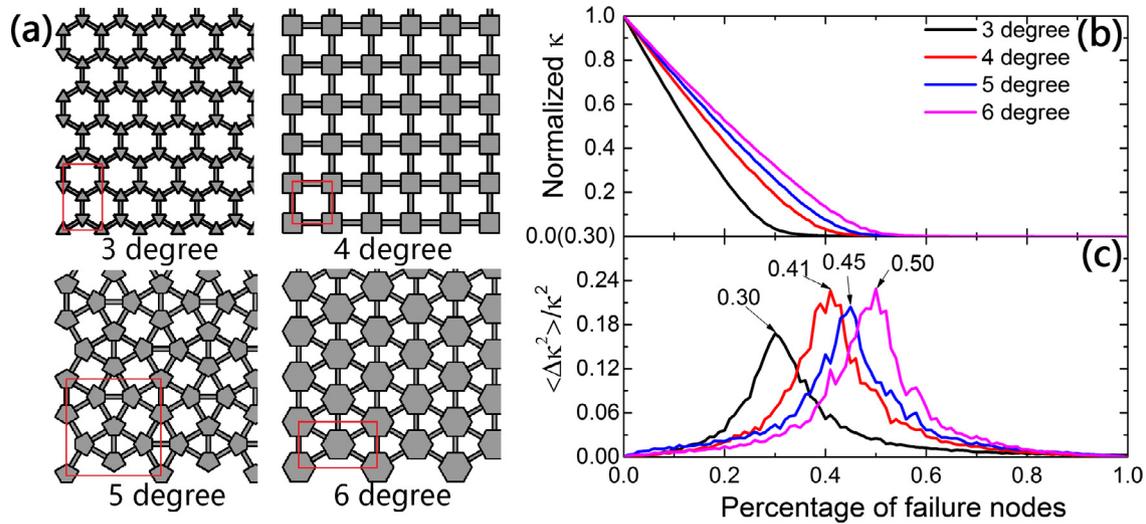


Fig. 2. (a) Structure scheme for 3,4,5,6-degree connected networks, in which the bond width is smaller than the corresponding edge length of nodes. The red rectangles stand for corresponding crystal cells. (b) and (c) are the corresponding normalized thermal conductivity and its fluctuation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

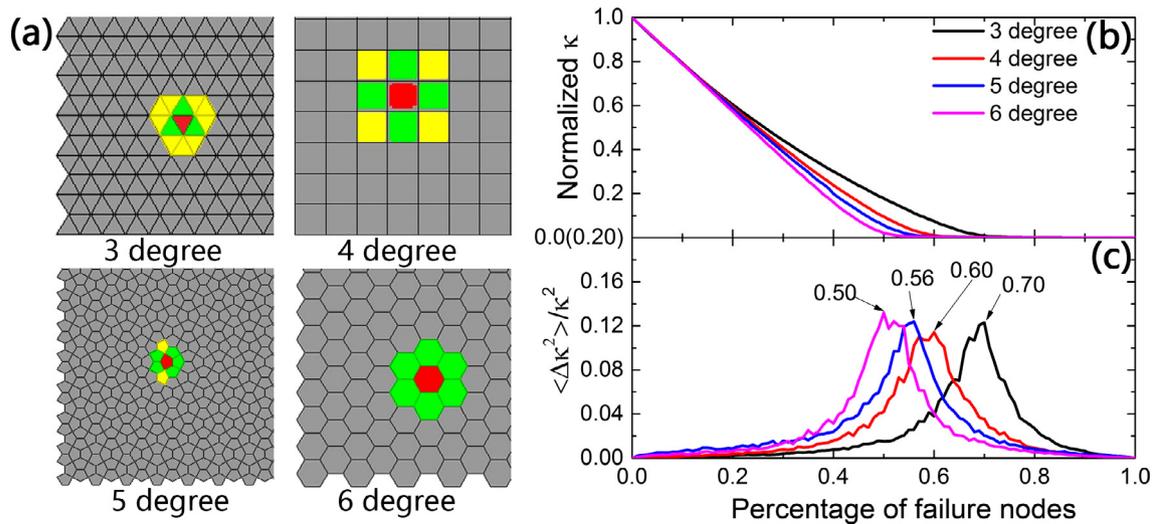


Fig. 3. (a) Structure scheme for 3,4,5,6-degree connected networks without bonds. (b) and (c) are the corresponding normalized thermal conductivity and its fluctuation.

[400 W/(m·K)]. We simulate 100 times for each percent of failure nodes, ranging from 0% to 100% with a step of 1%. All the effective thermal conductivities for each percentage are divided by κ_0 to get the ‘normalized κ ’ shown in Figs. 2–4 and 6, where κ_0 is the effective thermal conductivity of the network with no (0%) failure nodes. The normalized effective thermal conductivities and its relative fluctuations of these networks are shown in Fig. 2(b, c). As the connectivity increases, the stability (failure tolerance) of the network increases, and the corresponding critical point [peak of the curve in Fig. 2(c)] shifts to the right. This result is intuitive and in accordance with network theory qualitatively [22] since the robustness of the network increases with connectivity. Initially, we get Fig. 2(b, c) with bond length $L_{\text{bond}} = 0.2$ cm, bond width $W_{\text{bond}} = 0.05$ cm and bond conductivity $\kappa_{\text{bond}} = 4000$ W/(m·K). After changing one of these parameters and maintaining the other two, it is surprising to find that trends of the phase transition curves do not vary with bond lengths ($L_{\text{bond}} = 0.02, 0.04, 0.06, \dots$, and 0.20 cm), bond widths ($W_{\text{bond}} = 0.05, 0.10, 0.15, 0.19, 0.199, 0.1999$, and 0.19999 cm), or bond conductivities [$\kappa_{\text{bond}} = 10, 20, 50, 100, 200, 500, 1000, 2000$,

5000, and 10,000 W/(m·K)] as long as the width of bonds is smaller than the corresponding edge length of the nodes. We name this phenomenon as *bond-free property* for simplicity. It is worth noting that the number of crystal cells has no effect on the trends of the curves.

Fig. 3 shows similar results as Fig. 2. The only difference is that all the bonds are removed and the nodes are connected directly to each other. Although we have concluded previously that the trends of the curves do not vary with the bond length, the 3, 4, 5-degree connected networks switch to totally different trends when the bonds vanish. However, the curve of the 6-degree connected network remains unchanged. We can interpret this phenomenon from two different aspects. On one hand, these critical points agree with site percolation threshold for honeycomb lattices (6^3) [23,24], square lattices (4^4) [24], snub square puzzle lattices ($3^2, 4, 3, 4$) [23], and triangular lattices (3^6) [25]. They are special cases of the percolation theory. But this cannot explain the difference between Figs. 2 and 3. On the other hand, such phenomena can be explained by the transformation of connectivity. Taking

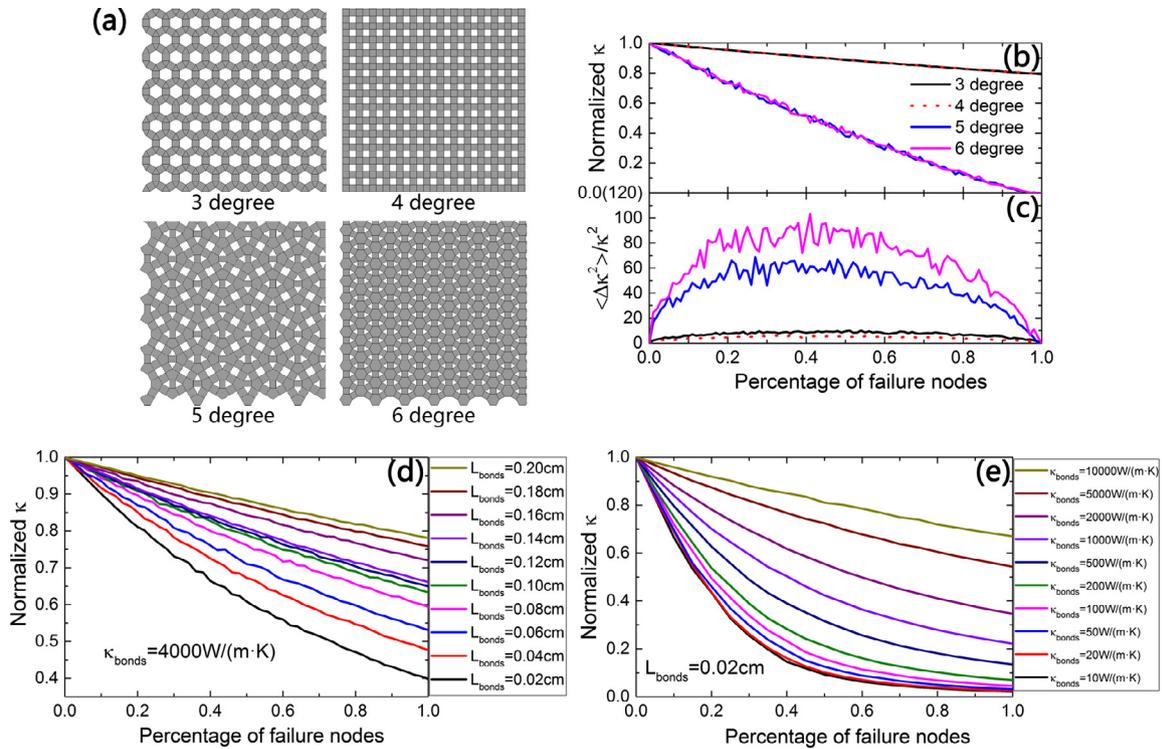


Fig. 4. (a) Structure scheme for 3,4,5,6-degree connected networks, in which the bond width equals the corresponding edge length of nodes. (b) and (c) are the corresponding normalized thermal conductivity and its fluctuation. Taking 3-degree connected network as an example, (d) and (e) show the relationship between the normalized thermal conductivity and the percentage of failure nodes for various thermal conductivities (κ_{bonds}) and bond lengths (L_{bonds}). The continuous variation indicates the validation of effective medium theory.

blue areas: perfect nodes or bonds $\kappa=400\text{W}/(\text{m}\cdot\text{K})$
 gray areas: failure nodes $\kappa=0.026\text{W}/(\text{m}\cdot\text{K})$
 red lines: one of the possible conductive channels
 green circle: interception of possible conductive channels
 green dots: single points connect intercepted channels

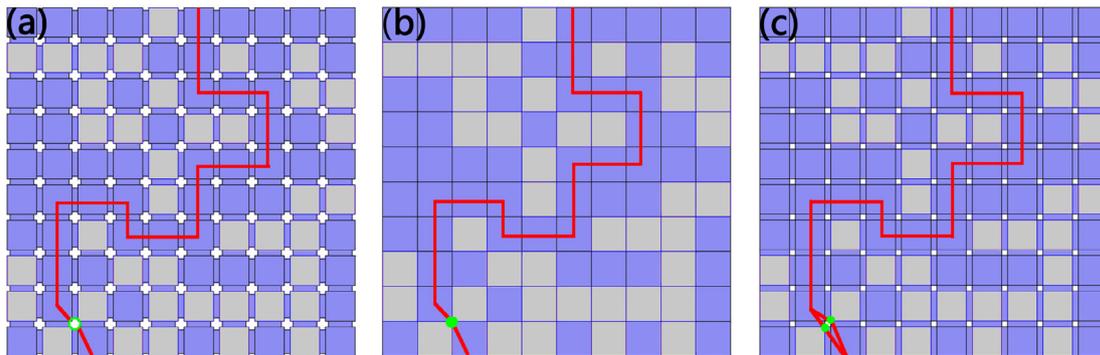


Fig. 5. Illustration of *single-point channels* taking 4-degree connected networks as example. Red lines stand for one of the possible path (conductive channel) for heat flux. (a) Network with width of bonds smaller than edge of nodes. The possible conductive channel is intercepted by empty spaces (white areas). More intuitively speaking, the red line is interrupted by the green circle. (b) Network without bonds. Intercepted conductive channel (red line) is connected again by the single-point connection (green dot) between the next-nearest-neighbor normal nodes. (c) Network with width of bonds equals edge length of nodes. Intercepted conductive channel (red line) is connected again by the single-point connections (green dots) formed by nearest neighbor bonds. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4-degree connected network as an example, as shown in Fig. 3(a), the red¹ node not only connects with the four green nodes, but also with the four yellow nodes via a single point. This single point can be regarded as a bond with infinitely small width and length according to the *bond-free property*. This point of view can be indicated by the

invariance of 6-degree connected network with/without bonds. So, in view of network theory, these 3,4,5-degree connected networks without bonds are actually 12,8,7-degree connected. While, the conventional network theory does not treat this single-point as a bond, so it may need further development. For convenience, we denote the channels containing single-point connection as *single-point channels* (see Fig. 5).

¹ For interpretation of color in Fig. 3, the reader is referred to the web version of this article.

This switching process from Figs. 2(b) to 3(b) is caused by the vanishing of bond lengths and the formation of *single-point channels* between nodes. We will discuss it deeply later [see Fig. 5(a, b)].

Now, let us focus on the condition that the bond width equals the corresponding edge length, see Fig. 4(a). The effective thermal conductivity and its fluctuation of the networks in Fig. 4(b, c) exhibit no phase transition and show totally different trends comparing to the previous two conditions. These networks do not conform with the *bond-free property*. Taking the 3-degree connected network as an example, Fig. 4(d, e) shows the trends of the normalized κ for various lengths and conductivities of bonds, which are obviously distinct from those shown in Fig. 2. The 4,5,6-degree connected networks show similar results. This continuous variation of normalized thermal conductivity can be explained by the well-known effective medium theory. That is, the effective thermal conductivity of the network is a composite made of four compositions: normal nodes [$\kappa = 400 \text{ W}/(\text{m} \cdot \text{K})$], failure nodes [$\kappa = 0.026 \text{ W}/(\text{m} \cdot \text{K})$], bonds [$\kappa = 4000$ or $10, \dots, 10,000 \text{ W}/(\text{m} \cdot \text{K})$] and empty spaces [$\kappa = 0 \text{ W}/(\text{m} \cdot \text{K})$]. It is worth noting that the duplicate of 3,4 and 5,6-degree networks in Fig. 4(b) are just coincidences, which can be indicated from Fig. 4(d, e) that the curve will shift upward or downward by changing κ_{bonds} and L_{bonds} . It will be discussed later why 5 and 6-degree connected networks turn to be thermal insulation while 3 and 4-degree connected networks do not.

3. Discussions

We have concluded the difference between Figs. 2(b) and 3(b) as a switching process in the previous section. It can be seen more intuitively from Fig. 5(a) and (b) that when the bonds length vanish, the network structure in Fig. 5(a) switches to the one in Fig. 5(b). A possible path [conductive channel, shown as red line in Fig. 5(a)] for heat flux that is intercepted by empty space [$\kappa = 0$, shown as green circle in Fig. 5(a)] can be connected by the single-point connection [shown as green dot in Fig. 5(b)]. Thanks to plenty of *single-point channels* formed by single-point connections, the κ_{eff} of the networks increase significantly and networks are more robust to node failures.

Similarly, the process of switching from Figs. 2(b) to 4(b) [from Fig. 5(a) to (c)] is due to the change of bond width, which comes

from the formation of *single-point channels* between bonds. Besides, the different properties of failure tolerance for networks shown in Figs. 3(b) and 4(b) [from Fig. 5(b) to (c)] can be regarded as the third switching process, which is due to both the disappearance of *single-point channels* between nodes and the formation of *single-point channels* between bonds.

Having known the significance of single-point connections and *single-point channels*, we are able to explain why 5 and 6-degree connected networks turn to be thermal insulation while 3 and 4-degree connected networks do not [in Fig. 4(b)].

For 3,4-degree networks, all the conductive channels formed by single-point connections (red dots) are still connected to the cold sources (blue lines) even if all the nodes fails [see Fig. 6(a, b)]. Thus, all the *single-point channels* formed by bonds will not be blocked by failure nodes. That's why the whole networks still remain pretty good thermal conductivity even all the nodes fails.

However, for 5,6-degree networks, these channels are truncated when the nodes which are directly connected to the cold sources break down, as shown in Fig. 6(c, d). Due to the role of these nodes directly connected to the cold sources [i.e. the nodes intersecting with dotted rectangles in Fig. 6(c) and (d)], the fluctuation of 5,6-degree networks are much larger, see Fig. 4(b, c). If the dotted rectangles in Fig. 6(c, d) are cut off and the red dots are connected to the cold sources directly [Fig. 6(e, f)], the networks still remain a good ability of conduction [see Fig. 6(g)].

4. Experimental verification

In order to verify the simulations based on finite-element methods, we conduct an experiment to confirm the above-mentioned three switching processes. The experimental samples are manufactured by chemical etching (main processes include gelatinizing, exposure and etching) using red copper [400 W/(m·K)], and they are covered with 0.01 mm-thick polydimethylsiloxane films in order to eliminate the infrared reflection. The experiment temperature profiles are obtained using FLIR E60 infrared camera. According to the simulation results, the effective thermal conductivity for the central area (black rectangle) shown in Fig. 7(b1, c1, d1) are 3.3, 43.0 and 151.1 W/(m·K), respectively. For the convenience of comparison, the background is etched with an array of holes to get an effective thermal conductivity of 151.1 W/(m·K). Despite the

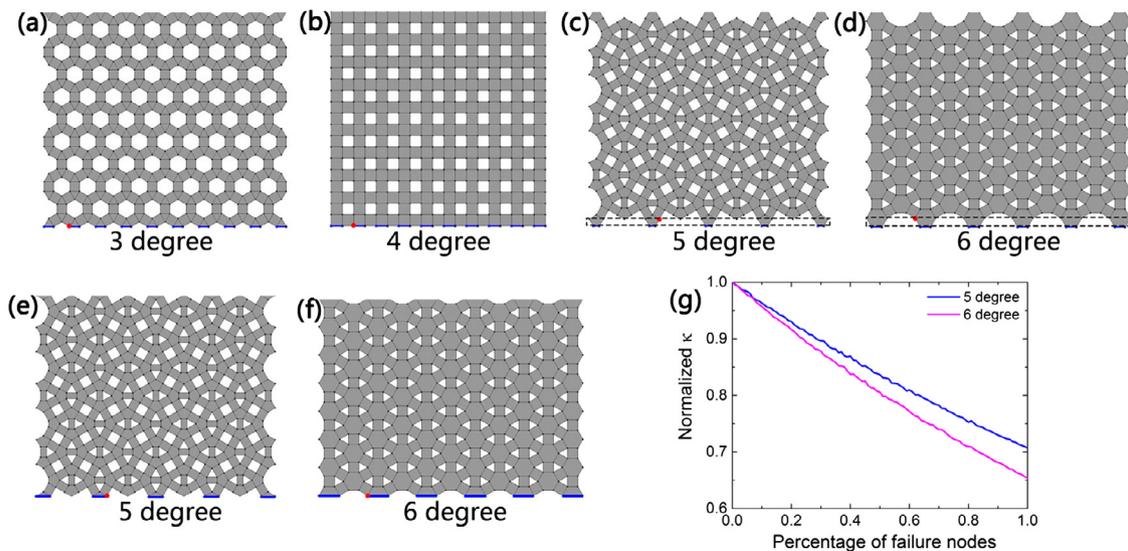


Fig. 6. Blue lines and red dots in (a)-(f) are respectively cold sources and single-point connections with the cold source. (g) shows the corresponding variation trends of the normalized thermal conductivity for the structures displayed in (e) and (f). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

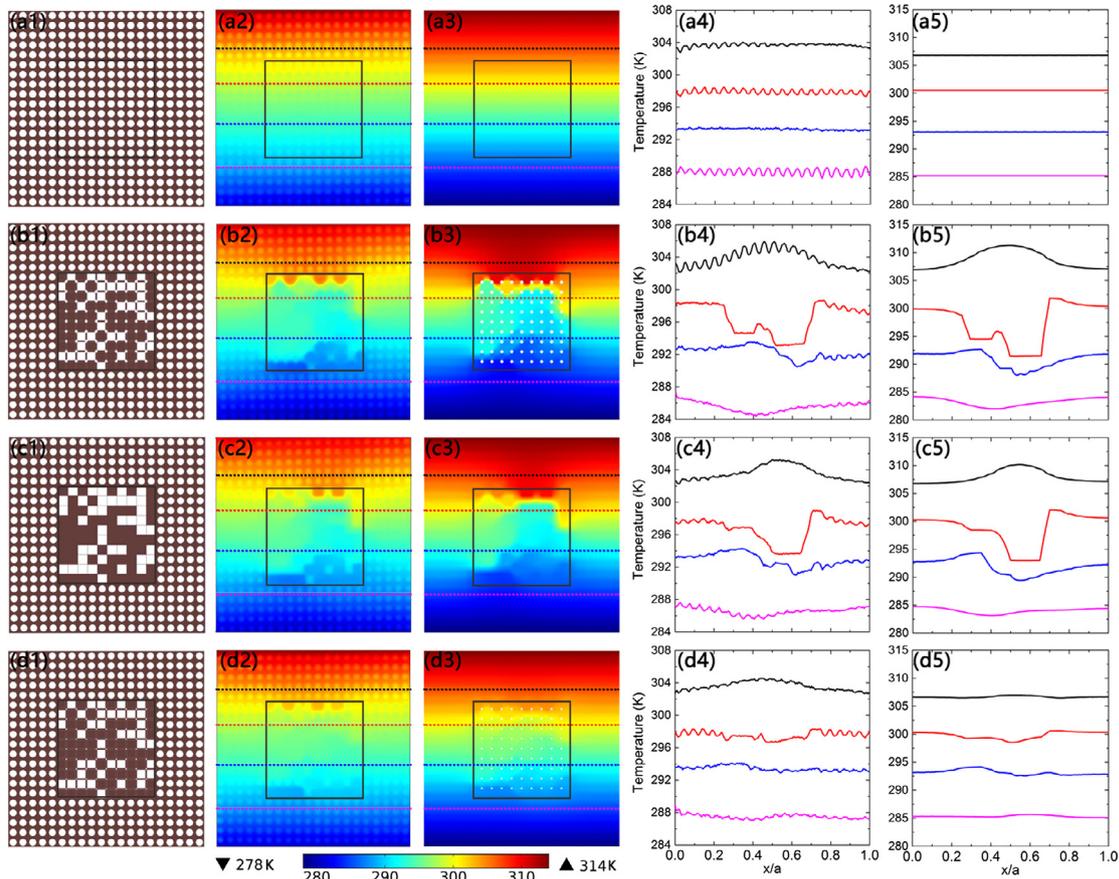


Fig. 7. Experimental verification of the switching process caused by geometric properties of the bonds. Structure schemes: (a1) background; (b1) 80% bond width (which means the bond width equals 80% of the corresponding edge length of nodes); (c1) without bonds and (d1) full bonds (which represents that the bond width equals the corresponding edge length). Dark purple regions shown in (a1)-(d1) denote red copper [$\kappa_{\text{redcopper}} = 400 \text{ W}/(\text{m} \cdot \text{K})$] and white regions denote air [$\kappa_{\text{air}} = 0.026 \text{ W}/(\text{m} \cdot \text{K})$]. (a2)-(d2) and (a3)-(d3) are experimental measurement and simulation results, respectively. The colorbar at the bottom indicates corresponding temperature distribution. (a4)-(d4) and (a5)-(d5) are temperature distributions along lines corresponding in (a2)-(d2) and (a3)-(d3). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

shaking of experimental temperature distribution in Fig. 7(a4)-(d4), the qualitative similarities between the experiment and simulation patterns (both the rainbow charts and line distributions) shows the reliability of the simulation results obtained in the previous section. The quantitative differences between experiments and simulations are caused by thermal convection of air.

5. Potential applications

Based on the switching process caused by bond width, here we propose two potential applications. One is thermal diode and the other may inspire researchers trying to improve thermal conductivity of polymer by formation of thermally conductivity networks.

Before explaining how the thermal diode works, we take a deep look at the switching process from Figs. 2(b) to 3(b) [i.e., from Fig. 5(a) to (c)].

Fig. 8 shows the effective thermal conductivity of the network versus the bond width for the 4-degree connected network with 40% of the nodes fail. The effective thermal conductivity of the networks experiences a 6 times jump [from 20 to 120 W/(m·K)] near the point at which the bond width equals the edge length of the nodes. This result is obvious and can be easily understood with the knowledge of single-point connection and switching process.

Using the same distribution of failure nodes as in Fig. 7, we can construct a network in which the bonds are made of material with positive thermal expansion coefficient [shown as the left panel of Fig. 9(a1, b1)] while the other with negative thermal expansion

coefficient [shown as the right panel of Fig. 9(a1, b1)]. After splicing them together, this structure can work as a thermal diode with high efficiency. Assuming that the diode works around 298 K, we can choose proper materials to realize such function: the 'left' bonds [namely, those located in the left part of Fig. 9(a1, b1)] expand to be wider than the node edges when the temperature is higher than 298 K and shrink to be narrower than the node edges when the temperature is lower than 298 K, and vice versa for the 'right' bonds. Thus, when we set the left boundary as the hot source and right as the cold source (defined as the positive direction of this diode), all the bonds expand and both parts transform to the kind of thermal networks shown in Fig. 4(a) that still remain a good ability of thermal conduction even though 40% of the nodes fail. That is, the diode works as on-state [see Fig. 9(a1, a2)]. While all the bonds shrink and both parts transform to the kind of thermal networks shown in Fig. 2(a), the networks experience a failure and the diode works as off-state [see Fig. 9(b1, b2)]. According to our simulations, this thermal diode has a large rectification ratio of 95.7%. Incidentally, although it is hard to find materials with appropriate thermal expansion coefficients and thermal conductivities, we can prepare the materials with the aid of shape-memory alloy, as proposed in Ref. [26].

Secondly, many researchers are looking for an efficient way to improve the properties of polymer by introducing networks into graphene. Zhang et al. have improved greatly the thermal conductivity of compounds by mixing with carbon nanocubes or graphene nanoplatelets [27]. Although such kind of networks are more likely

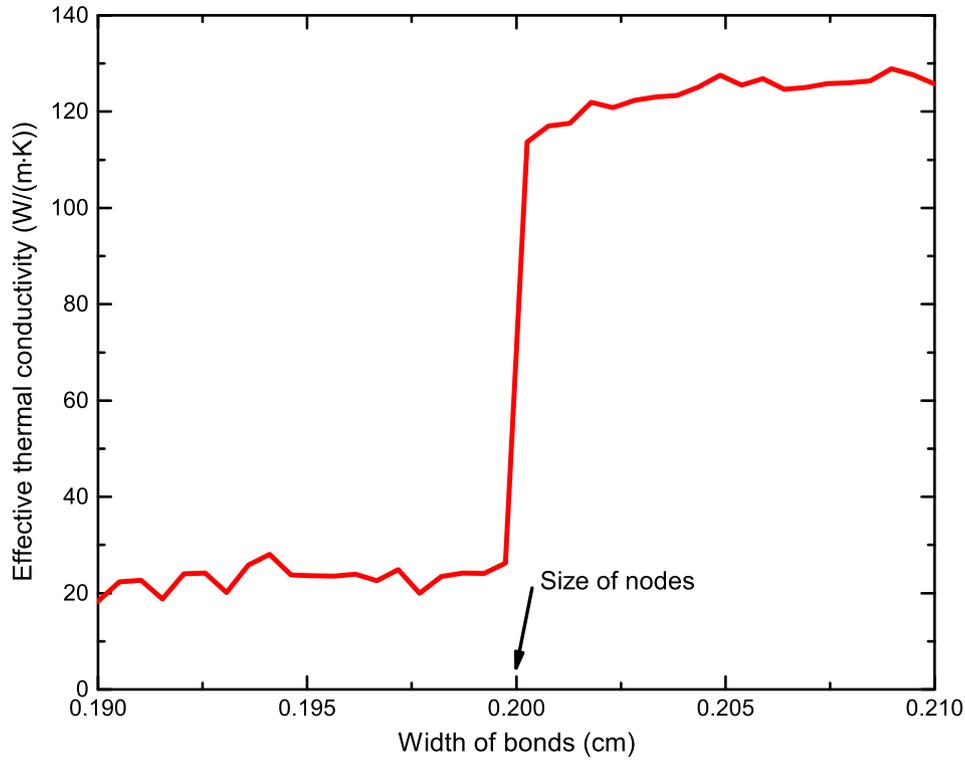


Fig. 8. Switching process caused by bond width. This simulation is performed for the 4-degree connected network with 40% of the nodes fail. Parameters: $L_{\text{bond}} = 0.02$ cm and $\kappa_{\text{bond}} = 400$ W/(m · K).

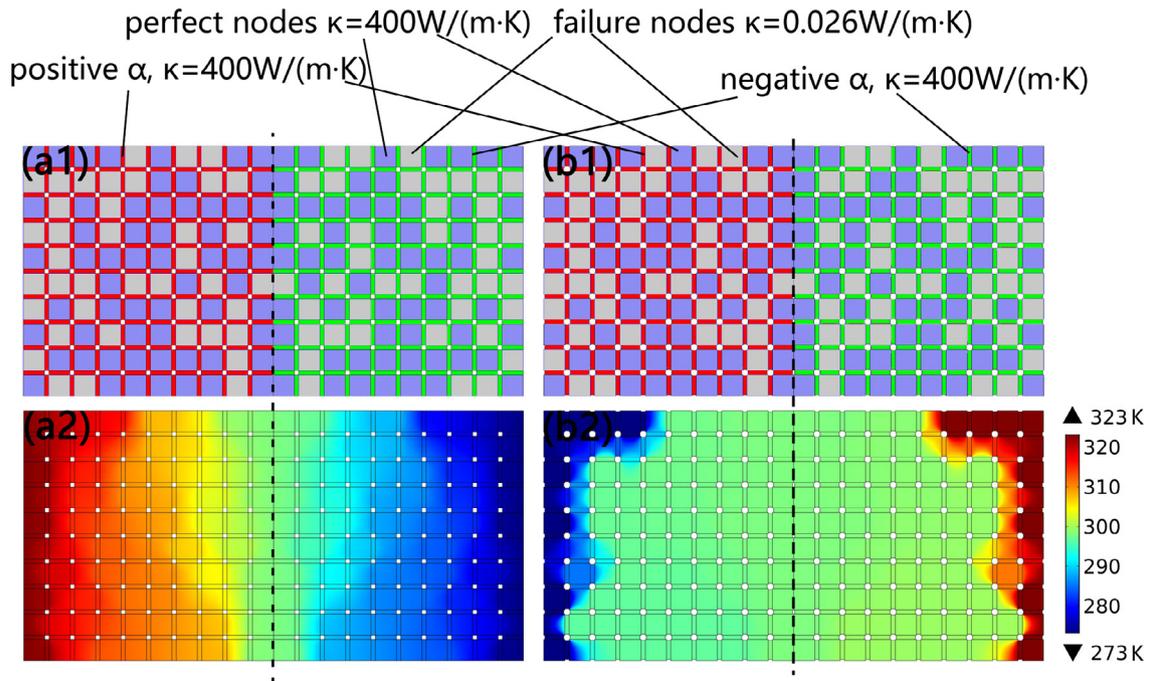


Fig. 9. Thermal diode based on the thermally conductive network. (a1) and (b1) are the diode structure designed for the on and off states, respectively. Red bonds on the left side of (a1, b1) are composed of material with positive thermal expansion coefficient α , while green bonds on the right side are composed of material with negative α . (a2) and (b2) are the corresponding simulation patterns of temperature distribution. This thermal diode has a rectification ratio $(J_H - J_L)/(J_H + J_L) = 95.7\%$, where high flux $J_H = 1.90 \times 10^5$ W/m² and (b1, b2) low flux $J_L = 4.12 \times 10^3$ W/m². The colorbar on the right side indicates temperature distribution of the diode. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

random networks, due to the conductive channels formed by wider bonds, it is reasonable to predict that the thermal conductivity may be improved greatly by choosing larger graphene nanoplatelets

than h-BN particles. This ideal is equivalent to using wider conductive bonds in our research, which can increase the thermal conductivity significantly.

6. Conclusion

In summary, we have investigated the connectivity and node influence of thermally conductive networks under three different conditions of bonds through finite-element simulations and reported the *bond-free properties*. The failure tolerance of these three different networks can be explained by network theory, percolation theory and effective medium theory, respectively. However, none of these theories can explain the discontinuous changes of κ_{eff} between the three kinds of networks. There seems to be certain incongruity among these theories. In order to reconcile the apparent incongruity, we propose three switching processes caused by geometric property of bonds. Based on the switching process caused by the change of bond width, we have also come up with two potential applications. It is worth mentioning that the results obtained in this work hold also for electrical conduction due to the analogue of mathematical formalism.

Conflict of interest

The authors declare that there are no conflicts of interest.

Acknowledgments

J.S. appreciates the useful discussion with Mr. Y.H. Gao. We acknowledge the financial support by the National Natural Science Foundation of China under Grant No. 11725521, and by the Science and Technology Commission of Shanghai Municipality under Grant No. 16ZR1445100.

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