



# Conductive compounds Temperature cools down

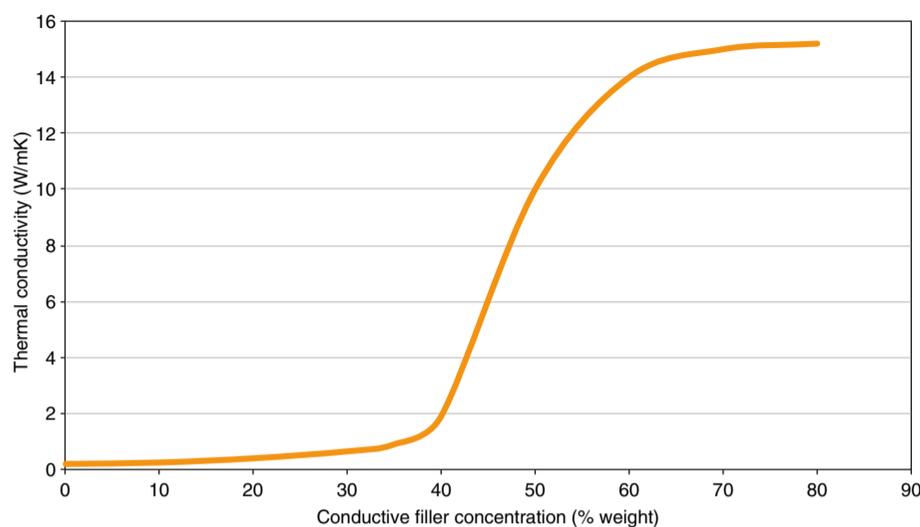
Between reality and false myths, here are some recommendations to take best advantage of the properties of thermally conductive compounds

**A**lmost ten years after their introduction to the market, the success of thermoplastic compounds with improved thermal conductivity can be considered as established.

Despite the inevitable initial mistrust and difficulties connected with the use of such materials, today many manufacturers of technical compounds include in their range at least one series of grades for injection moulding items requiring a better heat removal. However, the approach to these new proposals is not always correct, especially by users accustomed to historically proven materials, primarily aluminium or copper. So some “myths” have to be dispelled not to incur common design errors.



The black radiator of Tivedo lamp by Luceplan (designer Sebastian Bergne) realized with conductive compound Laticonther 62 G/50 by the Italian company Lati Industria Termoplastici 



**1** Qualitative curve of the percolation phenomenon in thermal conductivity. Below a certain threshold concentration level of the conductive filler the compound offers the thermal performance of the matrix only

### Main evaluation mistakes

A first mistake is to expect the same thermal performance of metals. This is a wrong assumption, as many successful applications showed that metals, in terms of heat exchange, are often oversized, and the same results can be reached by thermally conductive compounds with one-tenth thermal conductivity. This is especially true in the natural convection regime, i.e. when heat extraction is not carried out by forced ventilation. The second typical error is to continue to think about “standard” geometries – e.g. for heat sinks – i.e. designed for a good heat transfer, but always using metals. A new and more flexible mindset is to be entered into, which allows to understand that thermoplastic compounds are completely different materials, in particular with regard to their compositional inhomogeneities resulting in an physical anisotropic behaviour. Not considering this fact and treating plastics in the assumption that they behave like metals, may lead to significant design errors, sometimes ending in the failure of potentially viable projects. For this reason, it is extremely important that the most intimate properties of these materials are understood, “tamed” and capitalized on, especially in the design phase. In addition, requirements are not limited to thermal performance. There are, in fact, aes-

thetic constraints imposed by marketing and product designers, as well as electrical and mechanical properties. The development of increasingly efficient compounds even from this aspect, as well as the freedom of design, flexibility and efficiency of operation offered by these materials, are consolidating their increasing success in the most advanced technology sectors.

### Top yield at rock-bottom cost

If today it is safe to assume that the theoretical principles underlying the formulation and behavior of thermally conductive compounds have been defined and understood, the same cannot be said, however, for a wide range of technical and technological issues that are essential to ensure the fullest exploitation of the properties of these materials. Particularly during the design and manufacture of moulded items, in fact, the need emerges to ensure maximum yield at the lowest cost, i.e. the best thermal exchange with the lowest content of conductive filler. The success of the new applications in the lighting engineering, automotive or electric/electronic industry is achieved not only through the optimal formulation of compounds (choice of resin, filler, and corresponding proportions), but also, and especially, by optimizing the geometry of items and conversion techniques. The ideal framework should in-

## Measuring thermal conductivity

One of the instruments most commonly used and reliable for measuring thermal diffusivity ( $\alpha$ ) and therefore thermal conductivity, is the LFA by Netzsch. This instrument, by means of an IR detector, and through the emission of a flash generated by a xenon lamp, measures the time that the other side of a sample subjected to the flash takes to reach a constant temperature. Measurements can be carried out at different temperatures, as long as the following values are known, at those temperatures:

- $C_p$ , which can be obtained using a DSC for maximum reliability;
- density of the sample material ( $\rho$ ), which can be estimated by means of mathematical models at the different temperatures.

Once the diffusivity has been measured, applying the following correlation:

$$\alpha = \frac{k}{\rho C_p}$$

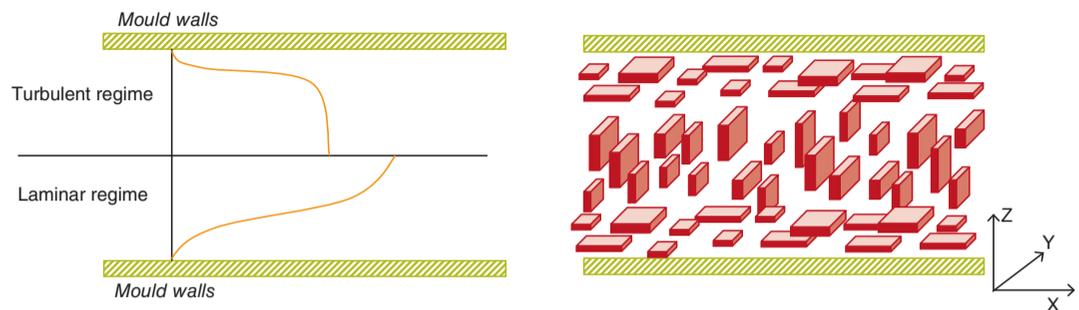
the thermal conductivity value (K) can be obtained.

clude an understanding of the relations between compound properties, product geometry, and moulding effects on heat transfer mechanisms. The choice of an inappropriate material, the non-exploitation of the real possibilities of compound or an unfavourable moulding results in a cost increase that may cause the failure of the project, unlike what happens to those able to work best on these fronts.

### Choosing the polymer

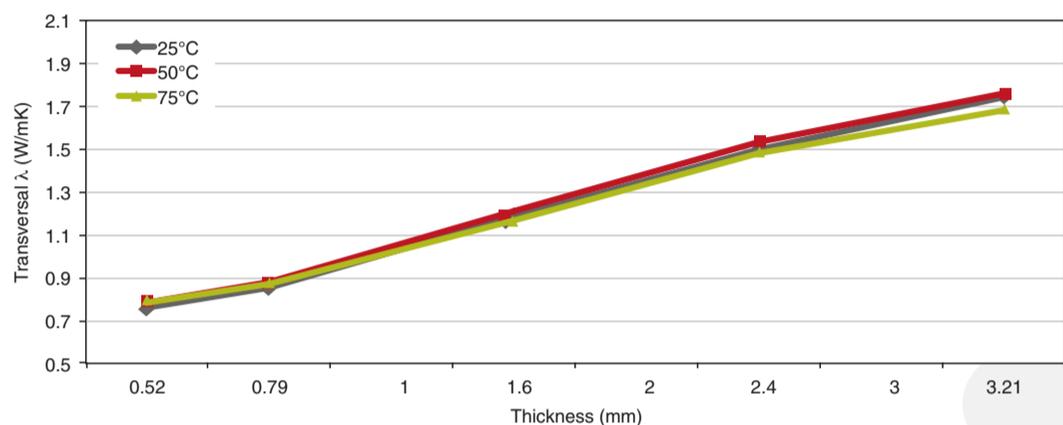
Under the thermally conductive solutions, the proposals ensuring maximum performance are obtained today by the extrusion of thermoplastic resins incorporating large amounts of fillers, such as PP, PC, PA6, PA12 or PPS, capable of transferring heat effectively. To this

**2** Qualitative curve of the orientation of fillers vs thickness: the thickness of the layer oriented along the direction of the melt that is measured at the mould walls depends on the speed profile; the more turbulent the motion of the melt is during the mould filling stage the smaller is the thickness



**Table 1** Form factor of certain fillers and their estimated conductivity

	ASPECT RATIO	K (W/mK)
Alumina	3.50	36.00
Magnesium oxide	3	23
Boron nitride	7	29
Graphite	4-6	75



regard, the most common choice is alumina or other salts and metallic oxides, graphite (if also a high electrical conductivity is acceptable), or hexagonal boron nitride (h-BN) when dielectric properties are required. As is well known, the most interesting results are obtained with graphite and boron nitride. In fact, these two fillers share not only interesting thermal properties, but also excellent processability, acceptable influence on the melt viscosity, and low abrasiveness, thus preserving extruders, presses, nozzles, and moulds from expensive and undesirable wear and tear phenomena. In any case, the amount of filler introduced into the matrix should necessarily allow to reach the percolation threshold (Figure 1), i.e. the condition in which contact occurs between the dispersed particles, associated with the occurrence of satisfactory thermal conductivity properties.

### Fillers and orientation effects

Most ceramic fillers, such as aluminium oxide, are composed of particles with geometric isotropy, i.e. having roughly the same extent in the three spatial di-

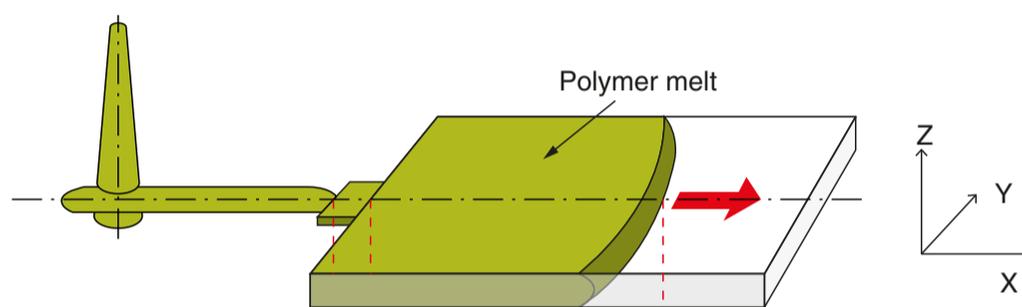
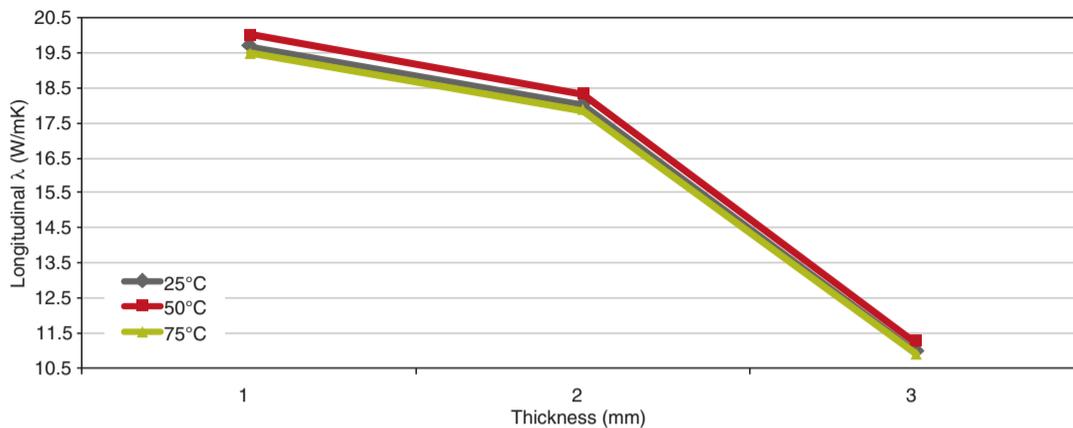
rections. So they are characterized by a form factor (or aspect ratio), understood as the relatively low relation between the larger and the smaller dimension. Instead, both graphite and hexagonal boron nitride (h-BN) show a high geometric anisotropy related to the communal crystalline morphology, an allotropic structure consisting of hexagonal systems arranged on stacked and independent sheets. Graphite and h-BN are available in flakes of different sizes and appearance, but all characterized by a quite high shape ratio (Table 1). The allotropic structure in which carbon atoms are arranged in graphite or boron, and nitrogen atoms in h-BN, results macroscopically in anisotropies also from the physical point of view: mechanical properties, as well as thermal conductivity, are extremely different depending on whether the direction is longitudinal or transverse to the planes.

### Filler properties

Due to their shape, first during extrusion and then during moulding, graphite and h-BN flakes are exposed to mechanical action generated by the shear stress in

**3** (top) and **4** (bottom) of the cross (z) and longitudinal (x, y) thermal conductivity as a function of thickness and temperature detected. The cross conductivity increases (z) along with the thickness of the wall filled. In general,  $k_x=k_y>k_z$  (bottom figure)

the flow of molten polymer. The distribution of these shear efforts is, in turn, influenced not only by the rheological properties of the compound, but also by the speed profile of material flowing into the mould cavity and the gradual cooling process occurring along the wall thickness to be filled. So also these flakes, as it happens for glass and carbon fibers, arrange themselves along different directions, as provided in different theories concerning the orientation tensor calculus. Typically, a high proportion of flakes oriented along the direction of melt flow near the mould walls is observed, as well as an orientation increasingly towards the center of the fluid vein (Figure 2). Since thermal conductivity of polymers of industrial interest is lower than that of graphite and h-BN by almost two orders of magnitude, it can be easily inferred that the capability of compound of transferring heat to the product is far higher



along the direction of maximum filler orientation than the transverse one (Figure 3). By performing measurements in the two directions of maximum orientation, e.g. using laser flash technique as provided by rule E1461, it can be observed that the thermal conductivity along the most favourable direction can even be five times higher than the normal one (Figure 4). It is therefore clear that the

design and engineering of devices for heat transfer cannot be separated from the understanding and control of this important aspect, which is linked both to the starting material properties – intended as a combination of resin and filler – and kinetics of cavity filling during moulding, as well as to the cavity geometry. The optimization of thermal conductivity of the product using the filler

orientation allows to minimize its content in the compound, thus reducing costs of the finished product, and to retain as much as possible the same resilience and elongation at break of the matrix, which are needed in the increasingly frequent case of inserts, self-tapping screws, and keying (Table 2).

### Effects of wall thickness

As mentioned above, theory and experimental techniques show a strong orientation of thermally conductive fillers along the direction of flow near the outer layers. This phenomenon leads to generally high conductivity values in the longitudinal direction; the thinner the filled walls, the higher these values. The random filler orientation does not occur in the transverse direction, as it might be assumed considering the typical shear stress conditions and the velocity gradient in the melt. A gradual orientation of particles in the perpendicular direction is instead observed, which results in a gradual increase of transverse thermal conductivity.

This first series of observations leads to two fundamental conclusions confirmed experimentally:

- the larger the thickness of the filled walls, the higher the integral thermal conductivity, intended as an average value over the product in the three spatial directions;
  - generally – and counterintuitively –, on average more effective thermally conductive items can be obtained if larger thicknesses are introduced into the areas expressly meant for heat transfer.
- It is clear that the factors influencing the shear stresses in the melt, too, are con-

**Table 2** Mechanical properties of the compound consisting of PA6 +50% graphite does not show such dramatic decline in performance as to prevent its use even in applications where an acceptable elongation at break is required

PROPERTIES OF THE MATERIAL	NORM	UNIT	PA 6	PA6 + 30% mineral filler (talc)	PA6 + 50% graphite	PA12 + 75% BN
Density in H <sub>2</sub> O	ISO 1183	g/cm <sup>3</sup>	1.18	1.37	1.50	2.24
<b>TENSILE STRENGTH</b>						
Tensile modulus		MPa	3,000	6,500	11,400	7,500
Stress at yield		MPa	50			
Elongation at yield	ISO 527-1	%	3.6			
Stress at break		MPa		70	60	47
Elongation at break		%	150	2.5	1.2	1.2
<b>IMPACT STRENGTH</b>						
Charpy with notch	ISO 179-1eA	kJ/m <sup>2</sup>	6	3	4	2
Charpy without notch	ISO 179-1eU	kJ/m <sup>2</sup>	NR	35	8	5

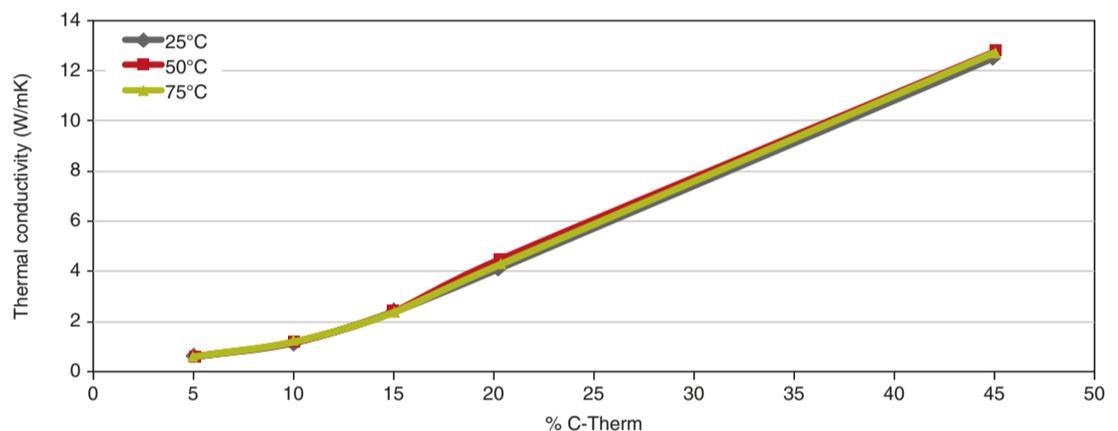
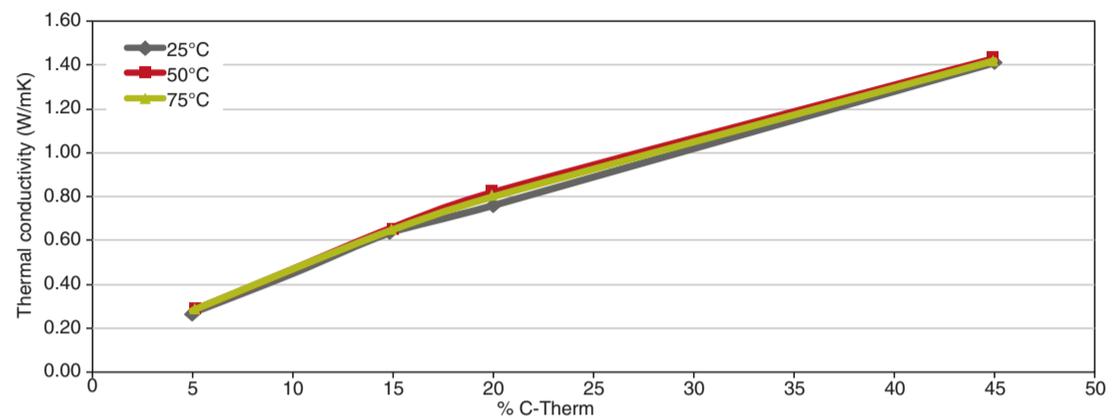
5 (top) and 6 (bottom) curve of cross and longitudinal thermal conductivity of PP in terms of C-Therm2 expanded graphite content

tributing to the filler orientation. These factors include, especially:

- filling speed: with increasing injection speed, a minimum increase in the longitudinal conductivity is observed;
- mould temperature: hotter moulds may induce a better transverse thermal conductivity thanks to the speed gradient reduction generated by the cooler external layers during filling.

### A few useful tips

Unfortunately, the cause-and-effect relationship among the factors governing the orientation of conductive fillers in compounds cannot be clearly and uniquely defined, mainly because most of the variables so far introduced are, in turn, linked to the compound composition. A major role is played, for example, by the thermal conductivity of material, which is a fundamental parameter for defining both melt viscosity and cooling rate. Based on what described so far, it is already possible to draw some valid technical measures for the production of thermally conductive plastic products. In fact, it can be inferred that the walls involved in a transverse heat flow – as in most heat sinks for lighting engineering – can be more effective if designed with a relatively large thickness. In fact, even if thermal resistance is theoretical-



ly higher compared to a thin wall, a larger thickness may offer an overall higher transverse thermal conductivity, making it more suitable for heat transfer across the external product surface.

### Process effects on filler

If it is correct to assume that the orientation of conductive fillers plays a fundamental role in obtaining good thermal properties, it is also true that the morphology of the particles plays an equally important role when these are dispersed in molten polymer, free to move and rotate under the action of shear stresses induced in the filling process. The shape, size and aspect ratio of boron nitride or graphite flakes will, in fact, determine their final set-up in the moulded product, and therefore the heat transfer capacity in the three spa-

tial directions. As can be inferred, larger flakes generally contribute to the increase in thermal conductivity due to the higher probability of contact with each other. For the same reason, especially structures that are as much as possible away from geometric isotropy are working well. Generally, the use of larger particles results in a substantial improvement in the longitudinal direction. However, this benefit is associated with smaller improvements in the transverse direction. In addition, it should be mentioned that these ceramics are made of extremely brittle particles dispersed in a melt with generally high viscosity. Both polymers and fillers are exposed to considerable mechanical stresses first by extrusion, and then by moulding processes, which inevitably result in a more or less accentuated



## Preparing the sample

The preparation of samples (of different geometries depending on the sample holder) is a delicate matter. The samples, with strictly constant and uniform thickness, are obtained by cutting printed larger specimens (it is very important to know the geometry and injection points of these samples in order to predict the orientation level of the fillers, which will affect the conductivity measured) which are then covered with a thin layer of graphite spray. When longitudinal thermal conductivity has to be measured, it is necessary to start from well-oriented samples that will be cut into "slices", rotated by 45° and put side by side.

rupture of brittle conductive flakes. The loss of thermal performance - especially in the transverse direction - is the immediate consequence of the reduction in size and aspect ratio of the particles. Some important conclusions can be drawn from these observations:

- geometry should also be considered in the selection of conductive fillers;
- with the same thermally conductive compound, conversion processes may have a major influence on the final product performance;
- to compensate for these inevitable difficulties, the compound manufacturer tends to add a certain fraction of excess filler. This decision determines benefits mainly as regards heat transfer in the transverse direction.

### Graphite like graphene

On this last point, some very interesting results have been obtained in the framework of an accurate study within the European project Nanomaster. The cross (Figure 5) and longitudinal (Figure 6) thermal conductivity of a compound obtained has been detected using a special graphite (C-Therm2 from Imerys) as a conductive filler, after subjecting it to a specific process in order to obtain the maximum dispersion and exfoliation effect. This resulted in the presence, inside the compound, of a variety of polydisperse nanometric flakes also including the single-layer structures known as graphene. This compound, which incorporates a conductive filler with a high form factor enhanced by the nanometric thickness

of these structures, made it possible to obtain percolation with very low filler concentration values. In this case too, the conductivity values measured grew in proportion with the amount of graphite used.

### Effects of moulding

Based on what has been defined so far, some simple guidelines for the configuration of the mould and the feeding system can be drawn. For example, too thin sections, which may induce high and prolonged mechanical stresses, should definitely be avoided. A good general rule, which is even more valid in the conversion of thermally conductive compounds, is to avoid too narrow runners and feed points, which are practical and inexpensive, but definitely unsuitable for brittle flakes of graphite or boron nitride. Also, much attention should be paid to the location of



weld lines, which are even more brittle in compounds reinforced with large amounts of anisotropic particles, as well as in particularly "cold" flow fronts. Instead, what cannot be inferred from the good moulding practice is the orientation of conductive fillers as a function of the location of the feed point, which is a new aspect with immediate effects on thermal conductivity, especially the transverse one. The arrangement of graphite or H-BN flakes is, in fact, dictated by the fluid dynamics of the molten compound, from the entrance into the mould to the completion of the structure. A random orientation in the immediate vicinity of the feed point is observed, which gradually becomes more regular, more and more driven by material flow (shearing flow).

In fact, it is in the areas farthest from the gate that the transverse thermal conductivity provides the highest values due to the high fraction of conductive fillers oriented perpendicularly to the direction of melt flow. This behavior may be thoroughly exploited by means of numerical simulations allowing to anticipate the arrangement of conductive particles as a function of filling kinetics and the local rheological behavior of the melt during mould filling. Correct computation requires, of course, simulation software providing the physical/mathematical description of a dispersed element with correct geometrical properties

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