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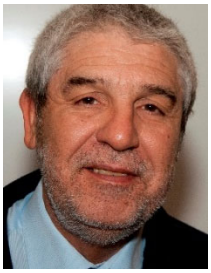

WHY ADDING FILLERS TO ADHESIVES? PROS AND CONS OF AN OLD BUT CURRENT PRACTICE IN ACADEMIA AND INDUSTRY

Fillers are solid materials with spherical, polygonal or fibrous geometrical shapes that can be added to adhesives/coatings/sealants for modifying their mechanical and/or rheological/viscoelastic properties, and/or decrease cost. Fillers can be of natural origin (sawdust, cellulose derivatives, minerals, etc) or synthetic (glass, fumed or colloidal silicas, graphene/graphite derivatives, etc).

Fillers are added to adhesives/coatings/sealants to impart different functions:

- Increase viscosity/modify the rheological/viscoelastic properties.
- Increase the mechanical properties (hardness, Young´s modulus, etc).
- Reduce/inhibit shrinkage during cure.
- Impart flame retardancy property.
- Increase the thermal stability.
- Impart colour.
- Control adhesion.

Several parameters determine the effectiveness of adding filler to adhesives/coatings/sealants, and they must be optimized to produce the requested performance.

1. Particle size. Particles of less than 5 μm are suggested. However, the addition of nanoparticles poses some drawbacks derived from different hydrophilic/hydrophobic balance and from the potential agglomeration of particles.
2. Particle size distribution. Narrow particle size distribution of filler particles is desirable to achieve a homogeneous distribution in the adhesives/coatings/sealants matrices.
3. Surface area. It determines the area of contact of each filler particle with the surrounded polymeric chains. Roughness should be differentiated from surface area because the addition of rough particles is not sufficiently efficient in controlling the viscoelastic properties of the adhesives/coatings/sealants.
4. Surface chemistry. In general, the surface of fillers is hydrophilic and the most of polymers in adhesives/coatings/sealants are hydrophobic. Therefore, in order to obtain an efficient miscibility of the fillers into the polymer matrices, their polarities must be balanced by surface modification of the fillers and/or the polymers in adhesives/coatings/sealants.
5. Aspect ratio. It is defined as the ratio between the length and the diameter of a filler particle. The larger the aspect ratio, the more available area for interacting the filler and the polymer. Thus, high aspect ratios are recommended in fillers (for example, platelets or fibres).
6. Sustainability concerns.

The main function of the fillers is disturbing the mobility of the polymeric chains in the adhesives/coatings/sealants causing physical cross-linkings. In fact, the degree of physical cross-linking increases by decreasing the particle size of the filler. The creation of chemical interactions between fillers and polymers is of great interest, but a competition between filler-filler and filler-polymer interactions can be faced.

Recently, several new issues related to the way to maximize the performance of fillers in adhesives/coatings/sealants have arisen. Among them, the method a mixing/adding the filler and adhesives/coatings/sealants has become critical in both solution and melted adhesives/coatings/sealants. Furthermore, the paradigm of “more added filler, better performance” is currently questioned. These two issues will be discussed in detail by focusing in the addition of different amounts of halloysite clay or silica to solid and solution polyurethane adhesives respectively.