# Effect of Filler–Fiber Interactions on Compressive Strength of Fly Ash and Short-Fiber Epoxy Composites

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**ABSTRACT:** Compressive properties of epoxy composites reinforced with fly ash and fibers, which have differing aspect ratios, are studied. Retention of strength and modulus are observed for a greater range of fiber volume fractions following fly ash introduction into the system. A slight decrease in density was also observed when fly ash content was higher, making these composites with materials of differing aspect ratio bearing reinforcement systems suitable in weight specific applications. The investigations showed that strength decrease is larger in fiber-bearing samples compared with only ash-bearing samples. This decrease was ascribed to the tendency of fibers to bunch. When the ash filler was introduced, this tendency of fibers to cluster appears to be reduced, resulting in increased strength and modulus. Further attempts are made to analyze these interactions of fibers and fillers through observations made on the surfaces of failed samples by scanning electron microscopy. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 87: 836–841, 2003

**Key words:** fillers; fibers; compression; scanning electron microscopy; composites

## INTRODUCTION

Fibers and fillers are generally used as reinforcements with polymers to improve their working properties. The properties of such composites are greatly influenced by the shape, size, and distribution of the reinforcing phase apart from its chemical composition and volume fraction. Fiber reinforcements with high aspect ratio are better utilized for improvement of strength and modulus required for aerospace and other high-performance applications, whereas the finer fillers with a far lower aspect ratio are included to improve specific properties in many lower-end applications.[1] Hence, there is a need for using such reinforcements with varying attributes in combinations to harness the possible synergistic effect that may result in widening the range of working properties.

Another important advantage that can be realized by combining these reinforcements is in their different rheological influences. Reinforcements possessing high aspect ratio render the mix highly viscous, whereas fine filler with low aspect ratio and wide range size distribution reduces the viscosity.[1] Hence, filler added to the mix of matrix polymer and short fibers not only improves the flow but also reduces the settling down of fibers. Bringing together of reinforcements can also reduce the amount of individual constituent materials needed to perform to specific levels in addition to resulting in a tangible cost reduction because of, for instance, the use of inexpensive material as one of the constituents. Also, optimum inclusion of the reinforcement combination can be achieved with a simultaneous reduction in the need for usage of a large amount of matrix polymer in the composite system.[2] Fly ash, a product of burning coal in thermal power plants, consists of fine spherical aluminosilicate particles with a wide range of size distribution. These attributes render fly ash a candidate material for freeinforcement in epoxy to realize cost effectiveness.

Fly ash has been used in earlier studies individually, as filler in thermoplastic[3] or thermoset[4,5] including epoxy[6] polymers, and in combination, with other fillers[2] and fibers.[7,8] But a focus on the filler–fiber interactions and especially on compressive properties has not been emphasized, as can be seen from a perusal of the available literature.

The present study, hence, addresses the issue of fiber–filler interaction when epoxy is compounded with short random fibers and fly ash particles. Compressive properties with various contents of fiber and filler introduced either individually or in combination are studied. Microscopic observation is undertaken to have a better perspective of interactions arising from differing aspect ratio bearing reinforcements in the epoxy matrix.

## EXPERIMENTAL

### Materials

The matrix system consists of a medium viscosity epoxy resin (LAPOX L-12) and a room temperature curing hardener with a tetra-amine functional group

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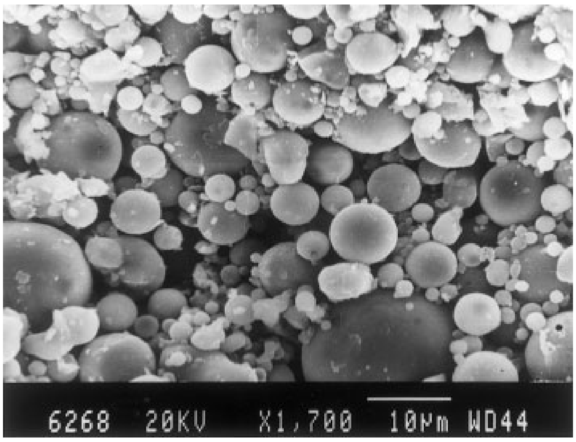


**Figure 1** SEM picture showing spherical fly ash particles of assorted sizes.

(K-6) supplied by ATUL India Ltd. The density of cured neat resin is 1120 kg/m$^3$. E-glass fibers, of density 2500 kg/m$^3$, treated with epoxy-compatible silane coupling agent, chopped to 6-mm length, and supplied by Fiber-Glass India Ltd., are used in the study. The filler used (i.e., fly ash) was obtained from Neyveli Lignite Corporation Ltd., Neyveli, India. This ASTM class 'C' fly ash with bulk density of ~900 kg/m$^3$ consists of a mixture of solid and hollow spheres of assorted sizes (Figure 1). Particle size distribution of this fly ash, as determined by the Malvern make laser light particle size analyzer, shown in Figure 2, consists of two distinct clusters emphasizing a gross bimodal nature. As per the bimodal theory of sphere–sphere packing, a distribution of the aforementioned nature should yield a packing value (82%) close to the theoretical maximum (85%).[9] Energy dispersive spectroscopy of the fly ash sample revealed the main constituents to be silica and alumina of ~63% and ~26%, respectively, whereas traces of other oxides, chiefly $Fe_2O_3$ (7%) and $TiO_2$ (2.5%), were also noticed. One of the objectives of using multiple components for reinforcing is to get the highest percent solid into the composite so that a significant reduction is realized in the amount of matrix polymer.[9] This objective can be achieved by using fibers with a higher *L/D* (i.e., aspect ratio) and filler of size such that *R* (the ratio of particle size and fiber diameter) is either small or large (>100).[9] Following this approach, in the present case, a fiber *L/D* of ~600 and a wide range ash particle sizes that are conducive for inclusion of a higher percent of solid in the composite were employed.

## Processing

A measured quantity of epoxy resin was mixed with a pre-weighed amount of fly ash and/or glass fiber, and the hardener was added to this with gentle stirring to avoid formation of air bubbles. The mixture was then slowly decanted into a mould (320 × 170 × 3 mm) coated before hand with uniform film of silicone-re-

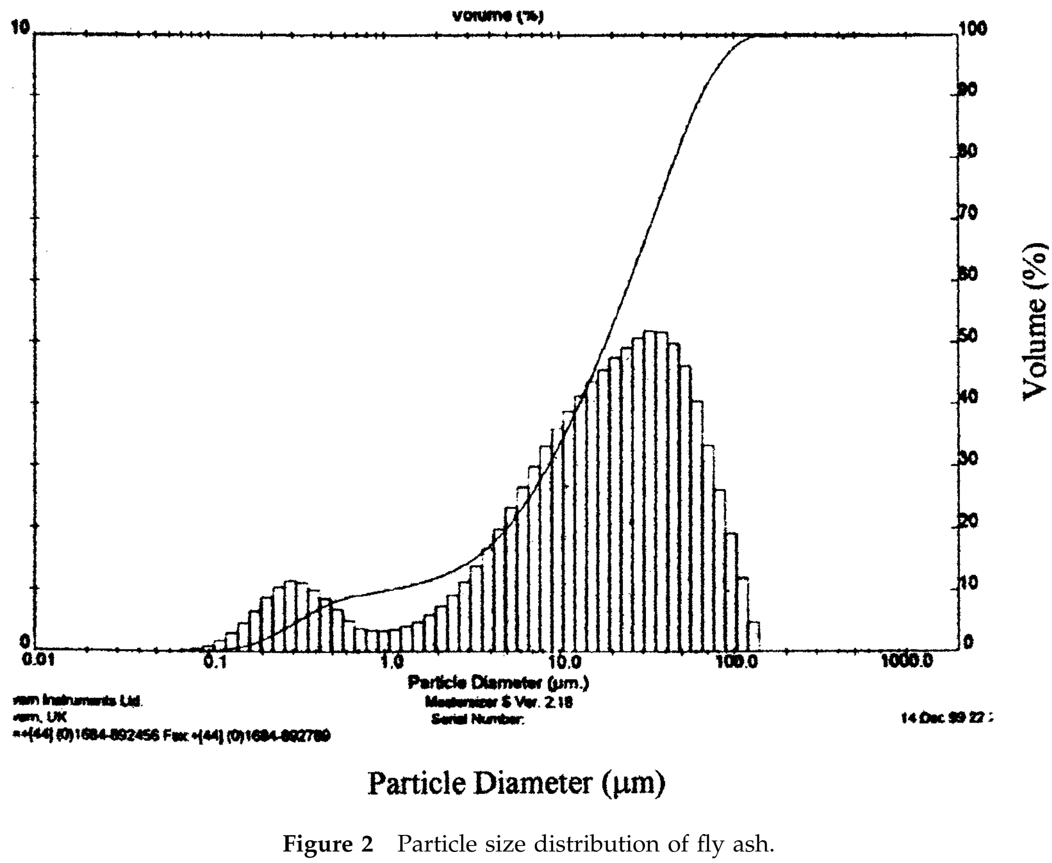


**Figure 2** Particle size distribution of fly ash.

**TABLE I**
**Table Showing Compositions of Systems and Densities**

| System | Filler (vol%) | Fiber (vol%) | Density (kg/m$^3$) |
|---|---|---|---|
| Neat Epoxy | — | — | 1120 |
| Fly ash | 5 | — | 1200 |
| | 10 | — | 1235 |
| | 15 | — | 1250 |
| | 20 | — | 1282 |
| | 25 | — | 1333 |
| | 30 | — | 1370 |
| Fiber | — | 5 | 1300 |
| | — | 10 | 1363 |
| | — | 15 | 1448 |
| | — | 20 | 1641 |
| | — | 25 | 1712 |
| | — | 30 | 1788 |
| Fly ash and fiber | 25 | 5 | 1328 |
| | 20 | 10 | 1375 |
| | 15 | 15 | 1452 |
| | 10 | 20 | 1476 |
| | 5 | 25 | 1489 |

leasing agent. The mixture that was like dough, especially for high volume fractions, was gently spread to fill the entire mould. The mould was then covered with a heavy lid with its underside having a Teflon sheet smeared with silicone-releasing agent. The mixture was left to cure at room temperature for ~24–26 h. Subsequently, post-curing was done at a temperature of 75°C for ~1.5 h. The cured rigid plate sample was withdrawn from the mould and edges were trimmed. In this way, epoxy-based systems with varying amounts of filler and/or fibers were cast (Table I). During the processing of samples containing both fly ash and fiber, their amounts are varied so as to maintain the total reinforcement at 30% by volume in the composite. Samples were then subjected to a 'C' scan nondestructive test to map out the regions of uniform material distribution. Test coupons of required size were then sectioned from such regions of the cast slabs.

### Compression testing

Compression testing was done in DARTEC 9500, a servo-hydraulic computer-controlled, testing machine. Test coupons (conforming to ASTM specification), of size 12.5 × 12.5 × 3 mm, were used for compression testing. The machine crosshead was programmed to apply the compression load at constant strain rate of 0.01 $s^{-1}$ throughout the entire duration of the test. From the load − stroke history, provided by the machine, the compressive modulii, and the strength were determined.

### Microscopic characterization

Samples subjected to compression were examined in a JEOL SEM (JSM 840A). The samples were gold coated in an ion sputtering unit before hand to make them conducting.

## RESULTS AND DISCUSSION

The strength and modulus for various volume fractions of fiber and fly ash introduced individually or in combination into the epoxy matrix are shown in Figures 3a and 3b, respectively. Strengths of the composites with only fiber show a tendency to record lower values at higher volume fractions. The reason for this declining tendency may be traced to what may be termed as either bundling or bunching together of fibers. Support for the occurrence of such an event is obtained when the SEM micrograph shown in Figure 4 is examined; that is, the top left-hand corner of the photograph has the ends of bunched fibers. The photograph (Figure 4) further shows (right and lower bottom of the picture) fairly clean fiber imprints where occasional debris is present. The point of significance is the clean form of the successive fiber depression showing fiber/matrix debonding due to interface separation. The situation is a direct result of poor spreading of resin on fiber because of lack of proper wetting of the surface of the clustered fibers during casting of the fiber only bearing composites. This bunching pro-

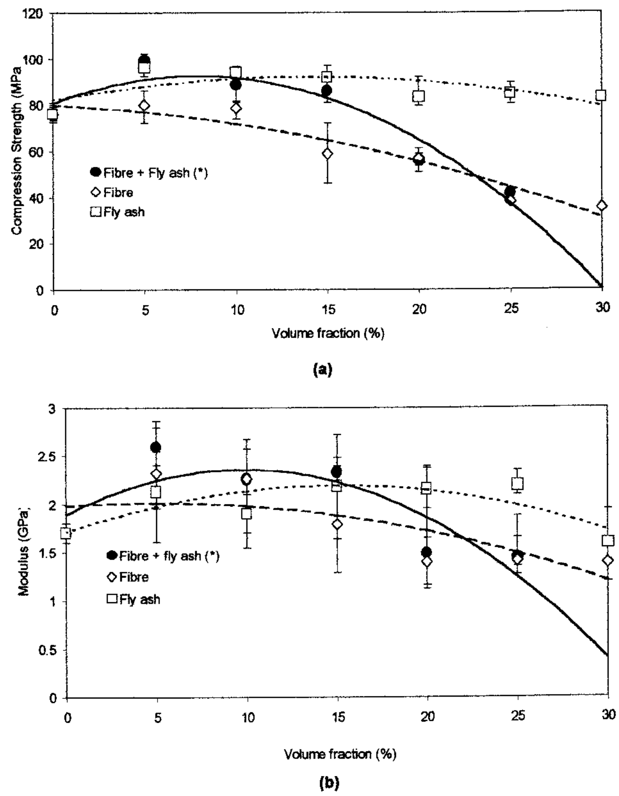


**Figure 3** Strength and modulus of short fiber and fly ash composites. The data on fiber + fly ash composites with changing fiber content, keeping the total volume fraction of the two constituent reinforcements at 30% by volume, are also included.

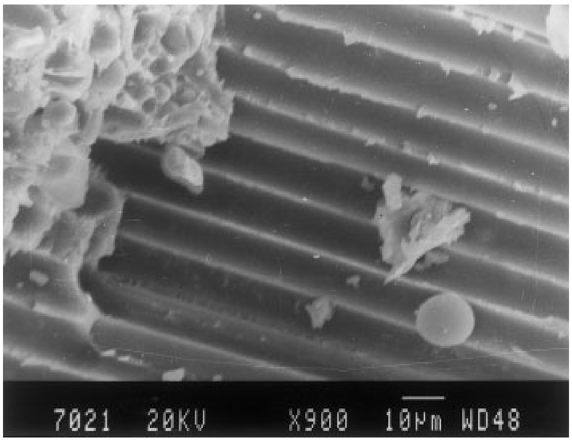


**Figure 4** SEM micrograph exhibiting the phenomenon of fiber bunching.

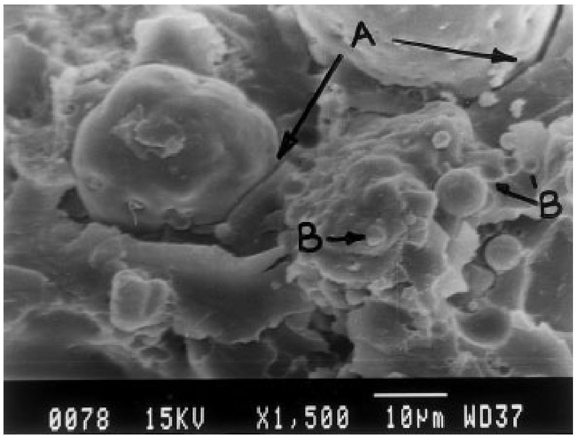


**Figure 6** Smaller ash particles adhering to the matrix and larger ones showing debonds.

cess leads to a reduction in the easily accessible surface area of fibers. Consequently, the wetting of the surface by the matrix resin, as emphasized earlier, is lowered. This point is also evident in Figure 5, where fibers with irregular adhesion to matrix are visible.

Following the initial raise, strengths of composites with only fly ash particles as fillers also show a decrease (Figure 3a), but this decrement is not as rapid as those recorded for composites with only fibers within them as reinforcement material. The possible reason for this lesser decrement in ash-bearing composites may be better wetting of filler particles, especially the smaller-sized ones, by the resin because bimodal particle distribution, referred to earlier, is involved in this work. The decrement noticed especially at larger volume percentages of filler ($>\sim$20–25%) may be traced to large particles, which are in great number at this volume fraction, being not wetted properly and carrying debonded surface around them. This observation is also supported by the application of Griffith's criterion for dewetting phenomenon[10] given by the expression, $\sigma_{\text{dewetting}} = Ar^{-2}$, where $\sigma_{\text{dewetting}}$ is the stress for dewetting. Larger particles show crescent-shaped debonds (shown by arrows at A in Figure 6) around them, which can be traced to the lower dewetting stress, as is evident in the equation just presented. The micrograph further reveals small particles adhering to the matrix (marked B). When the number of these larger particles increases at higher volume fractions, a decrease in the strength can be expected because these debonds around the larger particles act as stress raisers. Hence, a slight decrease in the strengths of fly ash composites is observed with increase in volume fraction.

When the filler is introduced in combination with fibers, the dispersion of the filler in the matrix might have first reduced the bunching and thereafter the proneness to insufficiently wet the fibers now with larger interfiber spacing. Figure 7 is an SEM picture taken on a matrix region at a place that is in between adjacent fibers. From this picture, good wetting characteristic displayed especially by smaller-sized particles [earlier illustrated (Figure 6) for filler-only-bearing composite] is apparent. This adhesion, together with the previously mentioned aspect of enhanced

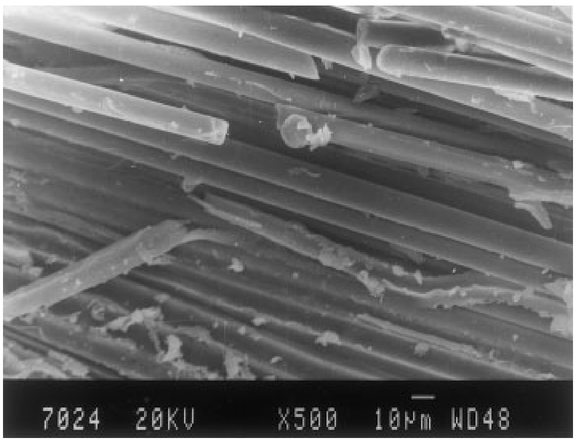


**Figure 5** A fiber-rich region showing irregular smear of resin on fiber.

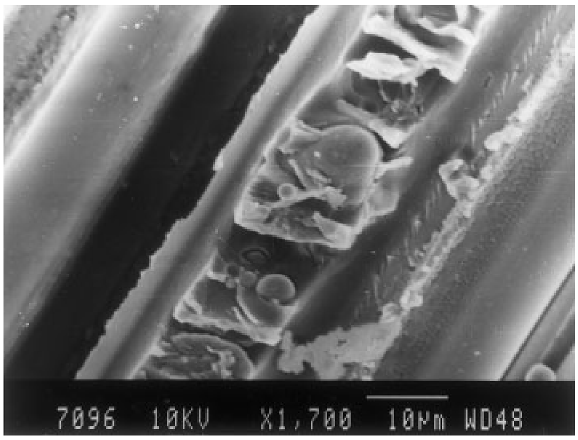


**Figure 7** The interfiber region occupied by ash particles.

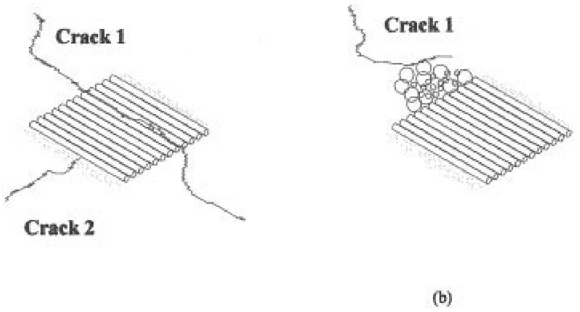


**Figure 8** Schematic showing the interaction of crack with fiber at two locations. It shows possible path taken by the crack due to occupying the edge of the fiber clusters.

interfiber spacing should have resulted in an improvement in strength (Figure 3a). The data (Figure 3a) show that incorporation of fly ash along with fibers has improved the retention of strength for a far greater range of fiber volume fractions. The variations of modulus of the composites for various volume fractions of filler, fiber, and their combination are depicted in Figure 3b. It can be noticed that modulus also is improved by the incorporation of filler with fiber.

The effect of strengthening due to addition of filler can be schematized, as shown in Figure 8. The fiber bundles, because of their high aspect ratio, introduce stress concentration at the ends and make the crack to take the path of least resistance (crack 1), which happens to be through the bundle because binding between the fibers is poor (Figure 8a). The crack, which fails to make it to such ends of the bundle and instead reaches them at the transverse section (crack 2, Figure 8a), could experience a resistance because of good adhesion of fiber to matrix due to the prior silane treatment given to fibers. How the situation at the ends of the bunch of fillers responds to the approaching crack when filler particles occupy such areas is a point needing consideration in this study of fiber–filler interactions. Here, because of the debonds (especially at larger-sized ones), the crack changes its path away from the one going through the bundle seen in the earlier case (Figure 8a). This situation is schematized in Figure 8b. Support for this argument is shown in Figure 9 where dispersed particles (shown by arrows), next to the fiber bundle (top left corner), enabling the fracture process to occur away from the bundle could be seen.

Another point of significance is that the densities increase with increased amounts of both fly ash and fiber steadily (Table I). Fly ash at 30% has a density of 1370 kg/m$^3$, and the corresponding value for the only-fiber-bearing composite is 1788 kg/m$^3$. The density values for the composites containing both filler and fiber vary between 1328 and 1489 kg/m$^3$. In this combination of individual materials, the fact that higher strength is recorded for larger volume of filler accompanied by lower volume of fiber is a point of considerable significance. Thus, 5% fiber + 25% ash, showing 99.7 MPa as compared to 42.1 MPa of 25% fiber + 5% ash, suggests that larger ash effectively helps in better distribution of fibers in the matrix. Also, when density values are considered (Table I), the higher fly ash (i.e., 25%) variety has a density of 1328 kg/m$^3$, whereas, 25% fiber bearing (in total of 30% introduced hybrid material) has 1489 kg/m$^3$. This simultaneous display of higher strength and lower density for larger ash-filler-bearing hybrid reinforcements can be used to an advantage when strength-to-weight considerations are important.

## CONCLUSIONS

The following points emerge from the investigation:

- The response of the epoxy system depends on the aspect ratio of the reinforcing medium under identical levels of introduced material.
- Fiber-only-bearing composite systems show a considerable decrease in strength, especially at large volume percentages. This decrease is less prominently seen in ash-bearing composites for identical volume levels of reinforcement.
- The fibers especially at large volume fractions display what is termed a 'bunching together' phenomenon. Consequently, the degree to which the resin can penetrate the interfiber region is reduced. This tendency being more at large volume fractions of fiber, the strength decreases due to this factor. For the ash-bearing situation, the better wettability of smaller particles account for the initial rise, and the debonds seen around large particles can be invoked for loss in strength at larger volume fractions of filler. Both these phenomena, namely the bunching (for the fiber case) and the differing adhesion factor seen depending on the size of the ash particle, are corroborated by scanning fractography.

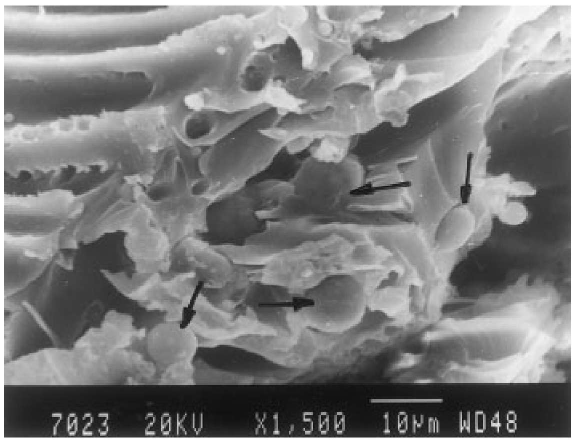


**Figure 9** SEM micrograph showing filler particles dispersed near the edge of a fiber bundle and showing the details of the path of fracture.

- The investigation demonstrates how the ash particles, especially at large volume fractions, are helpful in dispersing the fibers when the total volume fraction of the two material systems introduced into epoxy matrix is maintained constant. The resulting better strength of such a composite plus the lowered density and the cost due to the use of inexpensive fly ash makes this an economically and engineering viable system worth considering for development.

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