**1. Characterization**

Samples of the sedimentation experiment were prepared by the dispersions.

The viscosity test was acquired via a Brookfield viscometer (DV2T). According to the viscosity of samples, different rotor and speed was chosen. All the samples were tested at the same condition, such as temperature, humidity.

Scratch resistance test was obtained by using the five-finger scraper tester (The model of this machine is T113). The size of the specimen is 10cm\*8cm\*60μm. The speed of the tester was 10mm/s, and the total length was 10cm. The test conducted in different parts of the samples, starting with a small load, gradually increase the load. When the film of samples began to peel off, the corresponding pressure was recorded.

The adhesion test was conducted by using knife with six cutting edges and a blade spacing of 1mm or 2mm, special 3M adhesive tape with the brand name Scotch600. The samples were cut with uniform pressure, steady and the cutting speed was 20-50mm/S. Then rotate the samples 90 degrees and repeat the operation so as to form a grid pattern. A soft fur was applied to brush the diagonal lines on both sides of the lattice pattern, gently back 5 times, and forward 5 times. At last, the special 3M adhesive tape was used to test the adhesion. The test should be completed at least at three different locations of the specimen.

**2.Results and discussion**



Fig.1 The dispersion of G without LASS (a, f), G@LASS (10:1) (b, g), G@LASS (5:1) (c, h), G@LASS (2:1) (d, i), G@LASS (1:1) (e, j), G@LASS@Acrylic (k).

Fig.1 is the sedimentation experiment of G without LASS (a, f), G@LASS (10:1) (b, g), G@LASS (5:1) (c, h), G@LASS (2:1) (d, i), G@LASS (1:1) (e, j), G@LASS@Acrylic (k). The result of sedimentation experiment can evaluate the dispersion of G, G@LASS and G@LASS@Acrylic in aqueous solution. Sample (a), (b), (c), (d), (e) corresponds to the G without LASS, G@LASS (10:1), G@LASS (5:1), G@LASS (2:1), G@LASS (1:1), respectively. Sample (f), (g), (h), (i), (j) reflects the sample (a) after 12h, sample (b) and (c) after ten days, sample (d) and (e) after more than one month. Sample (k) is the composite of G@LASS@Acrylic after more than three months. From Fig.1, it is obvious that the G without LASS sank quicker to the bottom within 8h. What’s more, the G@LASS (10:1), G@LASS (5:1) also sank to the bottom after ten days. This was due to the little content of the LASS, the carboxyl groups of LASS chains were partly absorbed on the surface of G, which was not enough to stop the aggregation of the G. The G@LASS (2:1), G@LASS (1:1) could stay stable for more than one month. Therefore, the reaction between the G and LASS could truly improve the dispersity and stability of G. Then the weight ratio of 2:1 was chosen as the optimal ratio for G and LASS. The sample (k) in Fig.1 is the G@LASS@Acrylic composites, the composites could stay homogeneously for over three months. The effect of the LASS really enhanced the interfacial adhesion.

Table I. The apparent viscosity the samples

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Samples | Acrylic | Acrylic@LASS | | 0.5%wt G@LASS@Acrylic | 0.8%wt G@LASS@Acrylic | 1%wt G@LASS@Acrylic |
| Apparent Viscosity (mPa·s) | 82.25 | 82.25 | 82.28 | | 82.35 | 82.40 |

The Table I shows the results of the apparent viscosity of Acrylic, Acrylic@LASS, 0.5%wt G@LASS@Acrylic, 0.8%wt G@LASS@Acrylic, 1%wt G@LASS@Acrylic. The adding of G@LASS didn’t have a big influence on the viscosity of Acrylic. But the viscosity increased a little, because the adding of G@LASS introduced the hydroxyl and carboxyl which increased the number of ion charges in the system. As a result, the hydrodynamic volume of the colloidal particles and the motion resistance increased, which led to the increase of viscosity.

Fig.2 The scratch resistance of Acrylic (Sample 1), Acrylic@LASS (Sample 2), 0.5%wt G@Acrylic@LASS (Sample 3), 0.8%wt G@Acrylic@LASS (Sample 4), and 1%wt G@Acrylic@LASS (Sample 5).

The Fig.2 displays the scratch resistance of Acrylic, Acrylic@LASS, 0.5wt% G@LASS@Acrylic, 0.8%wt G@LASS@Acrylic, 1%wt G@LASS@Acrylic (which corresponds to sample 1, 2, 3, 4, 5, respectively). The minimum pressure of the Acrylic was 3N. When the pressure came to 3N, the film of Acrylic started to come off. According to the ISO-1518:1992 and GB/T 9279-2007, the minimum pressure can be used to reflect the scratch resistance of the samples. It was obvious that the scratch resistance increased significantly with the increase of Graphene. This reflected the adding of Graphene can improve the adhesion and the hardness of the coating, which resulted in the higher scratch resistance. It also corresponded to the results of adhesion of the samples. The G had a well dispersion in the Acrylic matrix. And the structure of Graphene wasn’t destroyed, so that the hardness of the composites was improved.

Fig.3 The adhesion test of Acrylic (a), Acrylic@LASS (b), 0.5%wt G@Acrylic@LASS (c), 0.8%wt G@Acrylic@LASS (d), and 1%wt G@Acrylic@LASS (e). (Due to the color of Acrylic and Acrylic@LASS coating are white, so the background is blank. In the contrary, the color of the rest is black, so the background is white.)

Fig.3 shows the adhesion test of the coatings. The adhesion of coating influences the quality of coating. The results of the test can be divided into five grades, corresponding to different standards. The specific method of rating is shown in the table II. According to the ISO-2409 and GB 9286-98, while the rating is first or second, the adhesion of the coating is regarded as qualified. The Fig.3 (a, b) corresponded to the Acrylic and Acrylic@LASS. It was obvious that they matched the second grades. The [0.5%wt G@LASS@Acryic](mailto:0.5%25G@LASS@Acryic), 0.8%wt G@LASS@Acryic and 1%wt G@LASS@Acryic was shown in the Fig.3 (c, d, e). It was clear that the adhesion of these coatings could reach the first grades. This indicated that the adding of G wouldn’t influence the adhesion of the Acrylic. In some degree, it could improve the adhesion of the Acrylic. It also reflects that G had a great interaction with the Acrylic matrix, so the excellent properties of G can also exhibit.

Table II. Grading method of coating

|  |  |  |
| --- | --- | --- |
| Grade/Level | Explanation | Surface appearance of cutting area |
| zero | The cutting edge is completely smooth without a lattice falling off. | 图片1 |
| first | There are a few coating shedding at the intersection, but the cross cutting area can’t be significantly greater than 5%. |
| second | There is a little coating shedding at the intersection of the incision, and the cross cutting area affected is obviously greater than 5%, but not significantly greater than 15%. |
| third | Coatings are partially or completely desquamated along the cutting edge, and/or partially or apparently pineapples at different parts of the cutting edge. The cross-cutting area affected is significantly greater than 15%, but not significantly greater than 35%. |
| fourth | The cutting edge of the coating is spalling off, and some or all of the squares appear to fall off. The cross cutting area affected is obviously greater than 35%, and it can’t be significantly larger than 65%. |
| fifth | The extent of spalling is over four. |