# Study on the process of Carbon Nanotubes modified Urea-Formaldehyde Resin Adhesive

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**Keywords:** Urea-formaldehyde, Carbon Nanotubes, Optimal solution

**Abstract.** Using mechanical blending of carbon nanotubes (CNTs) composite urea-formaldehyde (UF) resin adhesives. The ultrasonic time, the amount of CNTs, ultrasonic power orthogonal experiment was carried out. It is concluded that the important order of effect factors on free formaldehyde emission to UF resin adhesives: the amount of carbon nanotubes, ultrasonic time, ultrasonic frequency; The important order of effect factors on the shear strength of UF resin adhesives is ultrasonic power, the amount of CNTs, ultrasonic time; Preparation of composite CNTs/UF resin adhesives the optimal solution is: ultrasonic time of 30 min, ultrasonic power of 600 W, the amount of CNTs of 0.3 g.

## Introduction

In 1991, the Japanese scientists (Iijima)[1] for the first time discovered carbon nanotubes(CNTs). The chemical structure and atomic structure of CNTs are very simple, but there are very abundant physical and chemical properties. 1-2nm diameter of CNTs, young's modulus can reach 1 TPa [2], with diamond almost equal; The tensile strength of up to 150MPa[3]; The elongation of the CNTs can reach 20%, with good toughness; The strength of the CNTs is approximately 100 times of steel, and the density is only 1/6 of the steel. If CNTs were modified the other engineering materials, the new material strength, abrasion resistance and fatigue resistance performance has greatly increase[4].

Urea-formaldehyde (UF) resin adhesives have many advantages, such as technological process is simple and low cost, so man-made board with UF is widely used, but the downside is more obvious, the agglutination intensity is weak, the free formaldehyde emission is high, in order to improve this situation, Huang[5]etc with CNTs modified UF resin adhesives to improve the shear strength of UF resin adhesives and reduce the free formaldehyde emission. in this paper, by changing the ultrasonic time, content of CNTs and ultrasonic power, studying the orthogonal experiment, it is concluded the important order of effect factors on free formaldehyde emission to UF resin adhesives, the important order of effect factors on the shear strength of UF resin adhesives and the optimal solution of Preparation of composite CNTs/UF resin adhesives .

## Experimental

**The preparation of CNTs/UF resin adhesives.** The preparation of UF resin adhesive By Huang [5] methods. Different amount of CNTs added to UF resin adhesives, different ultrasonic time, different ultrasonic power.

**Determine the free formaldehyde emission of CNTs/UF resin adhesives** 5.00 g glue, add 30.00 ml of distilled water and 20.00 ml anhydrous ethanol, with 10% NH4Cl solution pH = 6.5, add 10.00 ml 10% NH4Cl solution, add 1.00 mol/l NaOH solution 10.00 ml, shake in full, in 20 ℃ for 30 minutes. In 1.00 mol/L hydrochloric acid standard solution modulation, until pH = 6.5. Twice the same test, record the data consumption of hydrochloric acid standard solution, take two test numerical average.

$$F = 0.03003 \times 6 \times (V1\text{-}V2) \times C/4 \text{ g} \times 100$$

F: the free formaldehyde emission (%);C: molar concentration of hydrochloric acid solution (mol/L); V1: blank consumption volume of hydrochloric acid (mL); V2: determination of test cost of hydroc hloric acid volume (mL); C(HCl)=1mol/L; G: sample quality (g).

**Shear strength test** With reference to GJB 1709-1993 adhesive shear strength test method at lo w temperature Sheet specimen material: the plank cypress, preliminary drying, drying to the moistu re content from 5% to 8%, will be tested in combination with surface planing, thickness of about 1-2mm. Specimen agglutination: use the beaker said take 10 g sample, add 1% ammonium chloride, a nd then mixing, mixing ammonium chloride fully into the adhesive. Shear strength test: use a ruler measuring on the surface of the specimen glue joint width and length (a) (b),bonding method as sho wn in figure 1 Shear strength calculation formula: F = p/(a ×b)
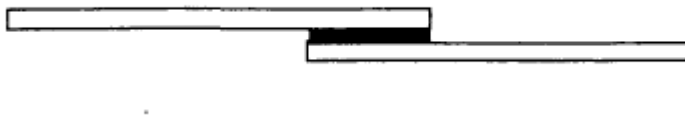


Fig. 1. Shear strength measurement

**CNTs/UF resin adhesives orthogonal experiment design** For ultrasonic time, the amount of C NTs and ultrasonic power three factors orthogonal design of experiment measuring shear strength a nd the quantity of free formaldehyde emission , orthogonal test are shown in table 1 (a) (b)

table 1 （a）

|  | A Ultrasonic time （min） | B Content of CNTs （g） | C Ultrasonic power （w） |
|---|---|---|---|
| 1 | 10 | 0.1 | 400 |
| 2 | 20 | 0.2 | 600 |
| 3 | 30 | 0.3 | 800 |

（b）

|  | A Ultrasonic time （min） | B Content of CNTs （g） | C Ultrasonic power （w） | Shear strength (Mpa） | Free formaldehyde （%） |
|---|---|---|---|---|---|
| 1 | 10 | 0.1 | 400 | 1 | 0.67 |
| 2 | 20 | 0.1 | 600 | 1.1 | 0.65 |
| 3 | 30 | 0.1 | 800 | 1.05 | 0.69 |
| 4 | 10 | 0.2 | 600 | 1.16 | 0.68 |
| 5 | 20 | 0.2 | 800 | 1.01 | 0.63 |
| 6 | 30 | 0.2 | 400 | 0.76 | 0.61 |
| 7 | 10 | 0.3 | 800 | 1.37 | 0.57 |
| 8 | 20 | 0.3 | 400 | 1.02 | 0.55 |
| 9 | 30 | 0.3 | 600 | 1.17 | 0.52 |

Table 2 for the ultrasonic time, the amount of CNTs, ultrasonic power of CNTs/UF resin adhesiv e composite free formaldehyde emission.

Table 3 for the ultrasonic time, the amount of CNTs, ultrasonic power of CNTs/UF resin adhesive shear strength of composite material.

| Table 2 | | | | |
|---|---|---|---|---|
| Variable | | A | B | C |
| The free formaldehyde emission | K1 | 1.92 | 2.01 | 1.83 |
| | K2 | 1.83 | 1.92 | 1.85 |
| | K3 | 1.82 | 1.64 | 1.89 |
| Range | R | 0.1 | 0.37 | 0.06 |
| Important order | | BAC | | |
| Optimal solution | | B3A3 C1 | | |

| Table 3 | | | | |
|---|---|---|---|---|
| Variable | | A | B | C |
| Shear strength | K1 | 3.53 | 3.15 | 2.78 |
| | K2 | 3.13 | 2.93 | 3.43 |
| | K3 | 2.98 | 3.56 | 3.43 |
| Range | R | 0.55 | 0.63 | 0.65 |
| Important order | | CBA | | |
| Optimal solution | | C2B3A1 或 C3B3A1 | | |

As is seen from table 2 that the order of range is:B: 0.37 > A:0.1 > C:0.06,showing that the important order of relevant factors that effect free formaldehyde emission of the CNTs/UF resin adhesives is: the amount of CNTs> ultrasonic time>ultrasonic frequency.

Determine the best solutions in each group by the size of the K1, K2, K3. Choose the minimum value in the A,B,C as optimal solution of free formaldehyde emission, the results is B: K3 = 1.64, A: K3 = 1.82, C: K1 = 0.83, showing that CNTs of 0.3 g ,ultrasonic time of 30 minutes, ultrasonic frequency of 400W is the optimal solution. Under the optimal solution, the free formaldehyde emission of CNTs/UF resin adhesive glue can be effectively  decreased.

As is seen from table 3 that the order of range is:C:0.65 > B:> A:0.63 0.55, determined by the range order of the ultrasonic time, the amount of CNTs, and ultrasonic power , showing that the important order of relevant factors that effect the shear strength of CNTs/UF resin adhesives is: C, B, A, namely: ultrasonic power, the amount of CNTs, ultrasonic time.

Determine the best solutions in each group by the size of the K1, K2, K3. Choose the maximum value of K in the A, B, C as optimal solution of shear strength performance, the results is C: K2=3.43, B: K2=3.56, A: K1=3.53 and C: K2= 3.43,B: K2=3.56, A: K1= 3.53, K2, K3, in group C the values of K2 and K3 are same, the power of K2 is 600W, the  power of K3 is 800W, based on the principle of lower consumption, we choose the power of 600W, just the K2, in other words, the optimal solution is the amount of CNTs of 0.3g , ultrasonic time of 30 minutes, and ultrasonic frequency of  600W. Under the optimal solution ,the shear strength of the CNTs/UF resin adhesive of adhesion agent is effectively improved.

Through the analysis above, we obtain two groups of optimum scheme. The optimal solution of the free formaldehyde emission is: CNTs of 0.3g,ultrasonic time of 30 minutes, ultrasonic frequency of 400W. The optimal solution of shear strength is:0.3 g for the amount of CNTs, ultrasonic time of 30 minutes, and ultrasonic frequency of 600W.As to the two groups of optimal solution, comprehensive balance method is used in analyze.

For A: ultrasonic time. Shear strength belongs to third main factors, free formaldehyde emission belong to the secondary factors, so choose the free formaldehyde emission, A3.

For B: the amount of CNTs. The largest of shear strength and free formaldehyde emission both are B3, so choose factors of B3.

For C: ultrasonic power. The effects of ultrasonic power on free formaldehyde emission is third, the  difference value is small, but for bonding strength, it is the primary factor, so C2 is selected.

Based on the analysis above,C2B3A3 is choosed as the. The optimal solution of the preparation of CNTs/UF resin glue adhesion agent is: ultrasonic time of 30 minutes, ultrasonic power of 600W, the amount of CNTs of 0.3 g.


**Conclusions**

In this study, CNTs was used to modified UF resin adhesives, realized the preparation of the CNTs/ UF resin glue adhesion agent, range is used to determine the important order of effect factors on fre

e formaldehyde emission to UF resin adhesives: the amount of carbon nanotubes, ultrasonic time, ultrasonic frequency; The important order of effect factors on the shear strength of UF resin adhesives is ultrasonic power, the amount of CNTs, ultrasonic time. Through value analysis of K in the orthogonal experiment, and comprehensive balance method to get the optimal solution is: ultrasonic time of 30 minutes, ultrasonic power of 600 W, the amount of CNTs of 0.3 g.

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