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# Optimization of carbon fibre production using the Taguchi method

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Since the variation of carbon fibre mechanical properties is very large, the Taguchi method was used to produce carbon fibre in a pilot plant, and to improve the properties of carbon fibre. Due to the production conditions of the Taguchi method, the tensile strength, modulus and breaking elongation of carbon fibre are increased by about 20, 13 and 26%, respectively.

## 1. Introduction

The quality of carbon fibres depends strongly on the composition and quality of the precursor fibres [1–6] and on the oxidation conditions [8–10], as well as on the carbonization conditions [11, 12]. Itaconic acid (IA) comonomer in polyacrylonitrile (PAN) fibre can depress the cyclized temperature, and 2-ethylhexyl acrylate (2-EHA) may prevent the cyclization propagation of the nitrile group during oxidation; but PAN precursor with a few percent 2-EHA comonomer has a preferred orientation [1]. When PAN precursor contains a larger side chain of acrylate comonomer, PAN precursor has a preferred crystal orientation and higher crystallinity, but the orientation of its resulting carbon fibre unexpectedly decreases [2]. The structures and mechanical properties for PAN precursor and its resulting carbon fibre were influenced by the distribution of comonomer composition among the chains [3]. In previous studies, PAN precursor with a composition of 98 mol % acrylonitrile (AN), 1.5 mol % 2-EHA and 0.5 mol % IA showed the best mechanical properties of carbon fibre [1]. It was also found as the aromatization index (AI) value for oxidized fibre is lower than 50%, the carbon fibre for a one-stage carbonizing furnace shows lower properties, but the properties can be improved by using a two-stage carbonizing furnace [8, 9]. An oxidized fibre with a lower AI value (60–70%) can be carbonized to form carbon fibre with higher breaking elongation, and the oxidized fibre with a higher AI value needs a shorter carbonization time [10]. If the oxidized fibre has an AI value of 60%, the carbon fibre carbonized at 800 °C for the first-stage furnace and at 1200 °C for the second-stage furnace exhibited better mechanical properties [11]. The higher the AI values of oxidized fibre, the larger the maximum stretching tension that can be applied at the first-stage carbonization. For oxidized fibres with the same AI value, the lower the carbonization temperature at the first-stage carbonization, the larger the maximum stretching tension [12].

Since the variation of mechanical properties for carbon fibre is very large, the excellent quality and reproductivity of carbon fibre are hardly obtained by using a single variable for investigating the production conditions. The variables include dependent and independent variables. Therefore, in this study, the Taguchi method was used to find the optimum production conditions, and to improve the properties of carbon fibres.

## 2. Experimental procedure

PAN copolymer was polymerized with acrylonitrile, methyl acrylate and itaconic acid in a mixed solvent of acetone and dimethylsulphoxide (DMSO) at 60 °C with α-α′-azo-bis(isobutyronitrile) as initiator under an inert nitrogen atmosphere. The resultant polymerization solution was directly spun to form PAN fibre using a spinneret (1200 holes, 0.06 mm hole$^{-1}$, length : diameter = 1) through a 25% DMSO coagulation bath at 30 °C then stretched in boiling water and dried. The properties of the PAN fibre are 1.2 denier per filament, 0.9 GPa tensile strength, 13.2 GPa modulus and 13.4% elongation. PAN fibres are stabilized in a continuous oxidizing furnace in air, then carbonized in two continuous carbonizing furnaces. The properties of a single filament of carbon fibre were determined by Vibrodyn for measuring fibre denier and Vibroskop (Lenzing AG) for measuring tensile strength, modulus and elongation with a testing speed of 1 mm min$^{-1}$ and a testing gauge of 10 mm. The modulus was measured at 0.5% elongation. The average mechanical properties of carbon fibre are calculated from the average value of 20 filaments in the tow of carbon fibre.

First design the experimental factors and their levels, and then create a three level column and replace the three level columns. From the experimental results, the signal noise (SN) ratio (larger the better) can be obtained from [13]

$$\mathrm{SN} = -10 \log \text{mean standard deviation (MSD)}$$

$$\mathrm{MSD} = 1/n \sum_{i=1}^{n} \frac{1}{y_i^2}$$

where $n$ is 20 for this study, and $y$ is the experimental data.

## 3. Results and discussion

Based on the authors' experiences in previous studies, the variables, affecting the properties of carbon fibre, and their levels can be selected and were set up as shown in Table I; then, these variables and levels were replaced as orthogonal arrays [14], as shown in Table II. Some levels of the variables for Nos 3, 8, 9, 12 and 14 in Table II were too high, which leads to easy breaking of carbon fibre during carbonization; therefore, these levels were lowered, as indicated by the numbers in parentheses. According to the experimental conditions in Table II, the tensile strength, modulus and breaking elongation of carbon fibres are obtained and listed in Table III. From the test data, the various SN ratios for the tensile strength, modulus and elongation of carbon fibres can also be calculated.

From the SN ratio in Table III, the average SN ratio of various variables, affecting the tensile strength of the carbon fibre, and their levels can be determined, as shown in Table IV and Fig. 1. Table IV shows that the most important two variables affecting the tensile strength of carbon fibre are the carbonization temperature during the first stage (with a contributive possibility of 27.4%) and the carbonization tension during the second stage (with a contributive possibility of 22.1%). If the carbonization temperature during the first stage is too high, the uncyclized molecules in oxidized PAN fibre with an aromatization index (AI value) or degree of oxidation of about 50–60% are easily degraded at higher temperature. Therefore, carbon fibre produced at 500 °C during the first-stage carbonization exhibits a good tensile strength. During the second stage carbonization, the larger the tension applied to the carbon fibre, the more preferred the crystal orientation for carbon fibre, and the better the tensile strength of the carbon fibre. Since the range of the oxidation temperature is very narrow, it only has a slight effect on the tensile strength of the carbon fibre. From Fig. 1 were selected the better levels of the various variables, such as A1, B1, C2, D1, E2, F1, G3 and H3 conditions, to produce the carbon fibre. The tensile strength, modulus and breaking elongation of the resultant carbon fibre are 4.1, $2.3 \times 10^2$ GPa and 1.6%, respectively. Comparison of the carbon fibre with sample No. 13 in Table III indicates that the tensile strength is increased by 20.6%.

From the SN ratio in Table III, the average SN ratio of the various variables, affecting the modulus of carbon fibre, and their levels can also be determined, as shown in Table V and Fig. 2. It is shown in Table V that the most important three variables affecting the modulus of carbon fibre are the carbonization speed (with a contributive possibility of 20.3%), oxidation time (contributive possibility, 17.4%) and carbonization tension during the second stage (contributive possibility, 15.9%). The slower the carbonization speed, which leads to be more perfect in the crystal for carbon fibre, the better the modulus of the carbon fibre. The oxidation time may affect the AI value of oxidized PAN fibre. Table V shows that 8 h oxidation time is the best for the modulus of the carbon fibre. The degree of oxidation of oxidized PAN fibre may influence the crystal and the crystal orientation for carbon fibre. The optimum conditions in Fig. 2 were used, such as A2, B2, C2, D2, E1, F3, E3 and H3 conditions, to produce carbon fibre. The tensile strength, modulus and breaking elongation of the resultant carbon fibre are 3.9, $2.6 \times 10^2$ GPa and 1.4%, respectively. Therefore, the modulus of the carbon fibre produced by the conditions of the Taguchi method can be increased by 13.0% compared to sample No. 11 in Table III.

TABLE II Experimental conditions

| Variable<br>Level<br>Run No. | A | B | C | D | E | F | G | H |
|---|---|---|---|---|---|---|---|---|
| 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 |
| 2 | 1 | 1 | 2 | 2 | 2 | 2 | 2 | 2 |
| 3 | 1 | 1 | 3 | 3 | 3 | 3 | 3(2) | 3(2) |
| 4 | 1 | 1 | 1 | 1 | 2 | 2 | 3 | 3 |
| 5 | 1 | 2 | 2 | 2 | 3 | 3 | 1 | 1 |
| 6 | 1 | 2 | 3 | 3 | 1 | 1 | 2 | 2 |
| 7 | 1 | 2 | 1 | 2 | 1 | 3 | 2 | 3 |
| 8 | 1 | 3 | 2 | 3 | 2 | 1 | 3(2) | 1 |
| 9 | 1 | 3 | 3 | 1 | 3 | 2 | 1 | 2(1) |
| 10 | 2 | 3 | 1 | 3 | 3 | 2 | 2 | 1 |
| 11 | 2 | 1 | 2 | 1 | 1 | 3 | 3 | 2 |
| 12 | 2 | 1 | 3 | 2 | 2 | 1 | 1 | 3(2) |
| 13 | 2 | 2 | 1 | 2 | 3 | 1 | 3 | 2 |
| 14 | 2 | 2 | 2 | 3 | 1 | 2 | 1 | 3(2) |
| 15 | 2 | 2 | 3 | 1 | 2 | 3 | 2 | 1 |
| 16 | 2 | 3 | 1 | 3 | 2 | 3 | 1 | 2 |
| 17 | 2 | 3 | 2 | 1 | 3 | 1 | 2 | 3 |
| 18 | 2 | 3 | 3 | 2 | 1 | 2 | 3 | 1 |

TABLE I Important experimental variables and their levels for producing carbon fibre

| Symbol | Variable | Level | | |
|---|---|---|---|---|
| | | 1 | 2 | 3 |
| A | Oxidation temperature (°C) | 220 | 225 | – |
| B | Oxidation time (h) | 7 | 8 | 9 |
| C | Oxidation tension (g f) | 800 | 1600 | 2400 |
| D | Carbonization temperature at first stage (°C) | 500 | 600 | 700 |
| E | Carbonization temperature at second stage (°C) | 1200 | 1250 | 1300 |
| F | Carbonization speed (m $\min^{-1}$) | 5.5 | 4.5 | 3.5 |
| G | Carbonization tension at first stage (g f) | 142 | 284 | 426 |
| H | Carbonization tension at second stage (g f) | 851 | 1277 | 1703 |

TABLE III Mechanical properties of carbon fibre and their SN ratio

| Run No. | Tensile strength (GPa) | SN ratio for tensile strength | Modulus ($\times 10^2$ GPa) | SN ratio for modulus | Elongation (%) | SN ratio for elongation |
|---|---|---|---|---|---|---|
| 1 | 3.0 | 8.9 | 1.9 | 44.8 | 1.5 | 2.4 |
| 2 | 2.9 | 8.3 | 2.2 | 46.5 | 1.2 | 0.8 |
| 3 | 2.2 | 5.7 | 1.9 | 45.3 | 1.1 | − 0.7 |
| 4 | 3.2 | 9.2 | 2.1 | 45.8 | 1.5 | 2.3 |
| 5 | 2.7 | 7.2 | 2.1 | 45.4 | 1.2 | 0.2 |
| 6 | 2.2 | 5.1 | 2.1 | 45.9 | 1.0 | − 1.4 |
| 7 | 3.2 | 9.4 | 2.2 | 46.7 | 1.3 | 2.2 |
| 8 | 2.7 | 7.5 | 1.8 | 43.4 | 1.3 | 1.2 |
| 9 | 2.8 | 8.4 | 1.8 | 44.1 | 1.4 | 1.9 |
| 10 | 2.1 | 3.9 | 1.8 | 43.8 | 0.9 | 0.1 |
| 11 | 2.9 | 8.5 | 2.3 | 47.3 | 1.2 | 0.5 |
| 12 | 3.1 | 8.2 | 1.7 | 44.2 | 1.3 | 1.0 |
| 13 | 3.4 | 9.7 | 2.0 | 45.4 | 1.4 | 2.0 |
| 14 | 2.5 | 6.1 | 1.8 | 45.0 | 1.1 | 0.7 |
| 15 | 3.0 | 8.2 | 2.1 | 46.2 | 1.3 | 0.1 |
| 16 | 2.6 | 6.6 | 2.1 | 46.2 | 1.1 | − 0.5 |
| 17 | 3.4 | 10.1 | 1.9 | 45.0 | 1.5 | 2.7 |
| 18 | 2.4 | 6.8 | 1.9 | 45.1 | 1.2 | 0.4 |

TABLE IV Assistant table for SN ratio of tensile strength

| Variable / SN / Level | A | B | C | D | E | F | G | H |
|---|---|---|---|---|---|---|---|---|
| 1 | 7.8 | 8.1 | 7.9 | 8.9 | 7.5 | 8.3 | 7.6 | 7.1 |
| 2 | 7.6 | 7.6 | 8.0 | 8.3 | 8.0 | 7.1 | 7.3 | 7.3 |
| 3 | – | 7.2 | 6.3 | 5.8 | 7.5 | 7.6 | 8.5 | 9.6 |
| Maximum–minimum | 0.2 | 0.9 | 1.7 | 3.1 | 0.5 | 1.2 | 1.2 | 2.5 |
| Contributive possibility (%) | 1.8 | 8.0 | 15.0 | 27.4 | 4.4 | 10.6 | 10.6 | 22.1 |

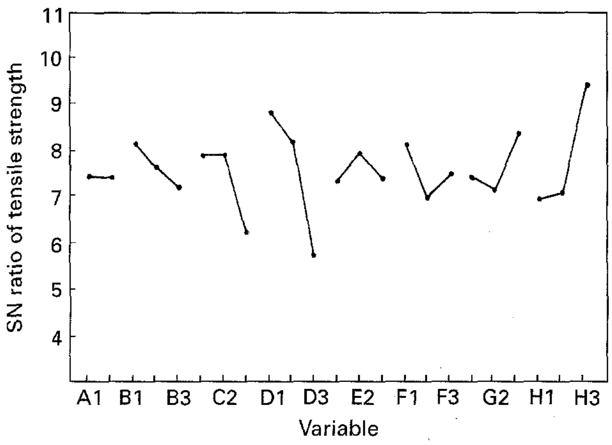


*Figure 1* Distribution of the SN ratio for the various variables affecting the tensile strength of carbon fibre.

The average SN ratio (Table VI and Fig. 3) of the various variables affecting the breaking elongation of carbon fibre, and their levels can be calculated from Table III. Table VI shows that the most important two variables affecting the elongation of carbon fibre are the carbonization tension at the second stage (contributive possibility, 27.6%) and the carbonization temperature during the first stage (contributive possibility, 23.7%). Since increasing the carbonization tension at the second stage leads to the development of more perfect crystals in the carbon fibre, and creates hardly any flaws or cracks in the fibre, the breaking elongation of the carbon fibre can be increased; because when the oxidized fibre is carbonized at a low temperature in the first carbonizing furnace, the uncyclized molecules of oxidized fibre are not easily

TABLE V Assistant table for SN ratio of modulus

| Variable / SN / Level | A | B | C | D | E | F | G | H |
|---|---|---|---|---|---|---|---|---|
| 1 | 45.3 | 45.6 | 45.5 | 45.5 | 45.8 | 44.8 | 44.9 | 44.7 |
| 2 | 45.4 | 45.8 | 45.4 | 45.6 | 45.4 | 45.0 | 45.4 | 45.7 |
| 3 | – | 44.6 | 45.1 | 44.9 | 44.8 | 46.2 | 45.9 | 45.8 |
| Maximum–minimum | 0.1 | 1.2 | 0.4 | 0.7 | 1.0 | 1.4 | 1.0 | 1.1 |
| Contributive possibility (%) | 1.5 | 17.4 | 5.8 | 10.1 | 14.5 | 20.3 | 14.5 | 15.9 |

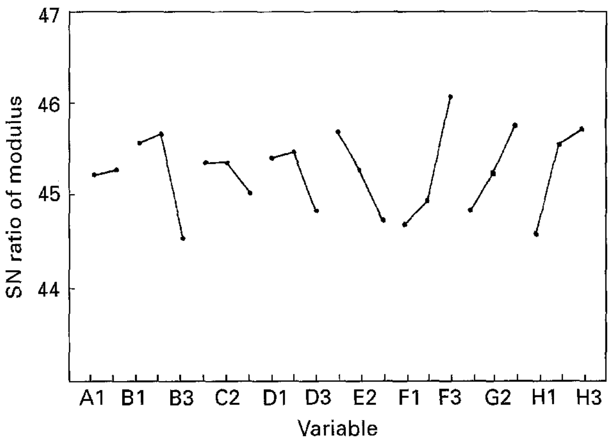

*Figure 2* Distribution of the SN ratio for the various variables affecting the modulus of carbon fibre.

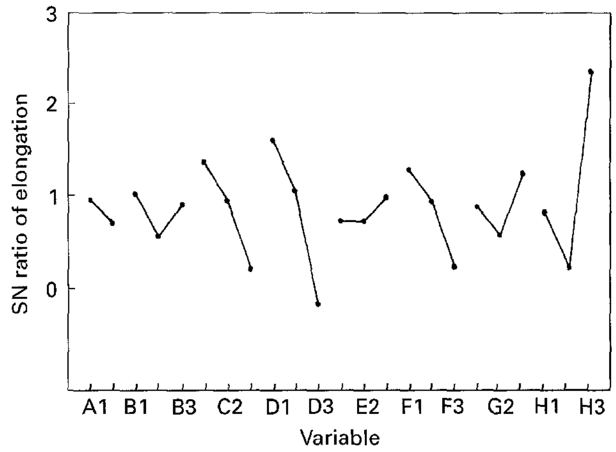

*Figure 3* Distribution of the SN ratio for the various variables affecting the elongation of carbon fibre.

TABLE VI Assistant table for SN ratio of elongation

| Variable / SN / Level | A | B | C | D | E | F | G | H |
|---|---|---|---|---|---|---|---|---|
| 1 | 1.0 | 1.1 | 1.4 | 1.7 | 0.8 | 1.3 | 0.9 | 0.9 |
| 2 | 0.8 | 0.6 | 1.0 | 1.1 | 0.8 | 1.0 | 0.6 | 0.3 |
| 3 | – | 1.0 | 0.3 | − 0.1 | 1.0 | 0.3 | 1.3 | 2.4 |
| Maximum–minimum | 0.2 | 0.5 | 1.1 | 1.8 | 0.2 | 1.0 | 0.7 | 2.1 |
| Contributive possibility (%) | 2.6 | 6.6 | 14.5 | 23.7 | 2.6 | 13.2 | 9.2 | 27.6 |

degraded, the elongation of its resultant carbon fibre is increased. From the above results, it was found that the most important variables affecting the elongation of carbon fibre are similar to those variables affecting the tensile strength of carbon fibre. From the optimum conditions, such as A1, B1, C1, D1, E3, F1, G3 and H3 conditions, (the level of the largest SN ratio) in Fig. 3, the tensile strength, modulus and breaking elongation of its resultant carbon fibre are 4.0, $2.1 \times 10^2$ GPA and 1.9%, respectively. The elongation of carbon fibre can be increased by 26.7% from the conditions of the Taguchi method compared to sample No. 17 in Table III.

From the above results, the various variables affect more strongly the breaking elongation and tensile strength of carbon fibre than the modulus of carbon fibre. The main factor affecting the modulus of carbon fibre is the carbonization temperature [15].

## 4. Conclusions

The most important variables affecting the tensile strength and breaking elongation of carbon fibre are the carbonization temperature during the first stage and the carbonization tension during the second stage; and the most important variables affecting the modulus of carbon fibre produced at about 1300 °C are the oxidation time, carbonization speed and carbonization tension during the second stage. From the conditions of the Taguchi method, the tensile strength, modulus and breaking elongation of carbon fibre can be increased by about 20, 13 and 26%, respectively.

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