Centrifugal Separation of Carbon Isotopes Using Carbon Tetrafluoride as Processing Gas

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Abstract. At present, the application of carbon isotope is becoming more and more extensive, especially in the field of medical testing, and the demand for $^{13}$C in high abundance is fast increasing. Using carbon tetrafluoride (CF$_4$) as separation medium, study on centrifugal separation of carbon isotopes was carried out. Single-centrifuge experiments were done based on modified domestic gas centrifuges. As a result, the separation factor and single-centrifuge separative power under different working conditions were obtained. Based on the results of single-centrifuge separation experiments, cascade calculation of the enrichment of $^{13}$C isotope was conducted by ideal cascade. According to calculation results, through two cascade separations, $^{13}$C isotope could be enriched from 30% to above 99% in abundance. The study laid a good basement for production of $^{13}$C isotope in high abundance.

1. Introduction
At present, the demand for stable isotopes in medical diagnostics, biopharmaceuticals, environmental science and other applications is increasing. The market has put forward higher requirement for product abundance of stable isotopes. Among them, carbon isotope is mainly used in the field of tracer. In the field of medical diagnosis, the high-abundance $^{13}$C isotope is mainly used for the detection of helicobacter pylori in the chemical form of urea. It has been demonstrated that the $^{13}$C can be increased from a natural abundance of 1.1% to more than 30% in abundance after three MARC cascade separations using heptafluoropropane (C$_3$HF$_7$) as medium for gas centrifugation [1]. In theory, heptafluoropropane can be converted to carbon tetrafluoride (CF$_4$). Based on this, centrifugal separation using carbon tetrafluoride as processing gas has been studied, which is aimed to increase the abundance of $^{13}$C from 30% to more than 99%.

2. Centrifugal separation medium
The separation medium of the gas centrifugation method generally satisfies the following requirements:
1) gas is stable at 300 °C without decomposition;
2) the relative molecular mass is not less than 70;
3) the saturated vapor pressure is not less than 665 Pa at normal temperature [2].

The relative molecular mass of natural CF$_4$ is 88.00. It is chemically stable, and the saturated vapor pressure is about 3.65 MPa at 15°C. Therefore, CF$_4$ meets all requirements.

Natural carbon has two stable isotopes, $^{12}$C and $^{13}$C, and their natural abundance is 98.89% and 1.11%, respectively. Natural fluorine has only one stable isotope, $^{19}$F. As a result, the centrifugal separation of carbon tetrafluoride is binary separation. The isotopic components of natural carbon tetrafluoride and their relative percentages are listed in table 1.
Table 1. Isotopic components of natural carbon tetrafluoride.

| Molecular composition | Relative molecular mass | Molar percentage (%) |
|-----------------------|-------------------------|----------------------|
| $^{13}$CF$_4$         | 89                      | 1.11                 |
| $^{12}$CF$_4$         | 88                      | 98.89                |

3. Single-centrifuge separation experiment

3.1. Experimental platform
In order to study the separation performance using carbon tetrafluoride as medium, a series of single-centrifuge separation experiments was carried out by the modified domestic gas centrifuge. The principle diagram of the experimental platform is shown in figure 1.

![Figure 1. Experimental platform schematic.](image)

The carbon tetrafluoride gas is passed to a modified domestic gas centrifuge at normal temperature. In order to ensure the stability of the feed stream, a stabilized container is connected in parallel with the feed bottle. After the separation by the centrifuge, two streams are obtained. The light component enriched stream is defined as the light fraction, and the heavy component enriched stream is defined as the heavy fraction. Because of the purpose of enriching the $^{13}$C isotope, the heavy fraction is product. Both fractions are collected by liquid nitrogen cold trap. The fluid parameters can be changed by adjusting the valve of the experimental platform. For different working conditions, the gas centrifuge needs to run continuously for one and a half hours under the rated rotating speed and power loss. Then the light and heavy fractions were sampled and analyzed.

3.2. Results and Analysis
According to the binary separation theory, the separation factor of carbon tetrafluoride is

$$q = \frac{C'_i}{(1 - C'_i)} / \frac{C_i}{(1 - C_i)}$$

In the formula, $q$ is separation factor, $C'_i$ and $C_i$ represent the molar percentage of the component having a relative molecular mass of 89 in the heavy fraction and the light fraction, respectively. The single-centrifuge experiments were carried out under different working conditions. The light and heavy fraction samples were analyzed by MAT-253 mass spectrometer, then data was calculated. The separation factor results are shown in table 2 and figure 2. It is inconvenient to list specific feed flow values, and qualitative analysis is carried out by feed pressure, which is linear with the feed flow.
It can be seen that when the cut is 0.5, as feed pressure increases, the separation factor remains basically unchanged first and then decreases. Feed pressure and separation factor are both important parameters to determine the separative performance of a single centrifuge, however, the trend of the two parameters is reversed. Therefore, the separative power is chosen to measure the separative performance, which takes the effect of the two parameters into account.

According to the condition of symmetric separation, the formula for separative power can be simplified as follow [3]:

$$\delta U = \frac{F}{2} \frac{q^{1/2} - 1}{q^{1/2} + 1} \ln q$$

In the formula, $\delta U$ is separative power, $F$ is feed flow. The separative power results are shown in table 3 and figure 3. The separative power is also inconvenient to list the specific values. $U_0$ is selected as a reference value. It can be seen that condition 3 is the best one under this experimental platform.

**Table 2. Separation factor results.**

| No. | Feed pressure (Pa) | Cut  | Separation factor |
|-----|--------------------|------|-------------------|
| 1   | 300                | 0.50 | 1.196             |
| 2   | 350                | 0.50 | 1.193             |
| 3   | 400                | 0.50 | 1.196             |
| 4   | 450                | 0.50 | 1.152             |
| 5   | 500                | 0.50 | 1.112             |

**Figure 2. Relationship between separation factor and feed pressure.**

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**Table 3. Separative power results.**

| No. | Feed pressure (Pa) | Cut  | Separation factor | Separative power ($U_0$) |
|-----|--------------------|------|-------------------|--------------------------|
| 1   | 300                | 0.50 | 1.196             | 0.68                     |
| 2   | 350                | 0.50 | 1.193             | 0.82                     |
| 3   | 400                | 0.50 | 1.196             | 1.00                     |
| 4   | 450                | 0.50 | 1.152             | 0.73                     |
| 5   | 500                | 0.50 | 1.112             | 0.47                     |
4. Cascading calculation

On the basis of single centrifuge separation results, the enrichment of $^{13}\text{C}$ was calculated by ideal cascade model. The working condition with the largest separative power is selected as the calculation parameter, that is, the feed pressure is 400 Pa, and the separation factor is 1.196.

Through two cascade separations, that is the heavy fraction of the first cascade is used as the feed of the second one, as shown in figure 4, $^{13}\text{C}$ isotope could be enriched from 30% to above 99% in abundance. The calculation results of the cascade parameters are listed in table 4 and 5, respectively.

![Figure 3. Relationship between separative power and feed pressure.](image)

![Figure 4. Schematic diagram of two separations.](image)

| Parameter   | No.1 | No.2 |
|-------------|------|------|
| Total stage | 52   | 70   |
| Feeding stage | 27   | 36   |

Table 4. Structure of cascade in two separations.
The abundance distribution of $^{13}$C in feed flow of each stage in two separations are shown in figure 5 and 6, respectively. The calculation results provide reference for the production of $^{13}$C isotope product.

### Table 5. Calculation results of cascade in two separations.

| No. | Fraction | Abundance of $^{13}$C (%) |
|-----|----------|---------------------------|
| 1   | $P_1$    | 81.45                     |
|     | $W_1$    | 3.68                      |
| 2   | $P_2$    | 99.02                     |
|     | $W_2$    | 14.90                     |

**Figure 5.** Abundance distribution of $^{13}$C in the first separation.

**Figure 6.** Abundance distribution of $^{13}$C in the second separation.
The gas centrifugal separation study was carried out with carbon tetrafluoride as the medium. The following conclusions can be obtained.

1) Separation of the $^{13}$C isotope can be achieved by gas centrifugation using carbon tetrafluoride as processing gas.
2) By comparing the separation factor and separative power, it is feasible to determine relatively great working condition. Under this experimental platform, the maximum of separation factor is 1.196.
3) $^{13}$C isotope could be enriched from 30% to above 99% in abundance by two cascade separations with 52 and 70 stages, respectively.

5. Conclusions and discussion

The gas centrifugal separation study was carried out with carbon tetrafluoride as the medium. The following conclusions can be obtained.

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References

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