Gasification Characteristics of Lignite in a Fluidized Bed Gasifier

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Abstract. Steam gasification experiment of lignite was carried out in a fluidized-bed gasifier. Effects of bed temperature, steam flow rate and particle size on the gasification behaviors were explored. Gasification behaviors of lignite with/without minerals were compared. When the bed temperature increased, the carbon conversion and gasification efficiency increased, H₂ and CO content increased, CO₂ content decreased, and CH₄ content kept essentially unchanged. When steam flow increased, the carbon conversion, gas yield and gasification efficiency increased, the content of H₂, CO and CO₂ in the syngas increased while the CH₄ content decreased. In this experiment, compared to the bed temperature and the steam flow, particle size showed only slightly influence on the gasification process. After removing the minerals, weight loss of lignite sample was almost same as the raw lignite, but the pyrolysis rate of the lignite with no minerals accelerated. In the experiment, although the carbon conversion kept no change, the content of H₂ and CO₂ decreased, CO content increased and CH₄ content fallen slightly. From the above, the inherent minerals in the lignite showed a positive influence on the gasification process, which improved the composition of the syngas.

Keywords: lignite, fluidized bed, steam, minerals, gasification

1. Introduction
Coal is the main energy in China, accounting for about 70% of primary energy consumption, and it is difficult to fundamentally change the energy consumption structure dominated by coal in the next few decades[1]. At the same time, about 80% of coal in China is used for direct combustion, and a large number of harmful gases and dust are emitted after combustion, which seriously damages the ecological environment. The fundamental way to reduce the environmental pollution caused by coal combustion is to develop and popularize the technology of coal optimal utilization, among which coal gasification is one of the main development directions[2]. Coal gasification technology is to convert solid coal into syngas, such as CO, H₂ and CH₄, which can be easily purified, and then used as fuel, preparation of chemicals or integrated coal gasification combined cycle power generation. The conversion of coal to syngas can significantly improve resource utilization efficiency, effectively control pollutant emissions,
and provide convenience for subsequent conversion and utilization[3-4].

On the other hand, lignite is a kind of coal with low degree of coalification in coal resources, and lignite in China is a kind of inferior fuel with high volatile matter, high moisture, high ash content, low calorific value and low ash melting point[5]. If the lignite is used to combust in direct, the high moisture and ash content of lignite make the lower boiler thermal efficiency and greater dust and greenhouse gas; high moisture also increases the transportation cost. Compared with other utilization methods, lignite gasification has the following advantages: (1) Lignite has good reactivity and high carbon conversion because of its high volatile content; (2) the volatile matter and moisture of lignite help to improve the gasification environment, especially the water can be used as an important source of hydrogen in the gasification process; (3) the gasification products are easy to be purified, transported and utilized. Therefore, lignite gasification is an ideal utilization mode at present[6].

In this paper, lignite was used as experimental material and steam as gasifying agent to study the gasification characteristics of lignite. The influence of raw material properties and operating conditions on gasification process was explored, and the influence of minerals in coal on gasification characteristics was preliminarily explored, so as to provide basic data and reference for fluidized bed lignite gasification process.

2. Experimental part

2.1 Experimental materials

In this experiment, lignite in Yunnan Xiaolongtan was selected as experimental material, and three groups of pulverized coal with different particle sizes were used: 0.3mm-0.38mm (fine particles), 0.5mm-0.6mm (medium particles), and 0.7mm-1.0mm (coarse particles). The industrial analysis and element analysis of raw coal as received basis are shown in Table 1.

| Proximate analysis wad/% | M    | A    | V    | FC   |
|-------------------------|------|------|------|------|
|                         | 15.46| 21.03| 38.72| 24.79|

| Ultimate analysis wad/% | C    | H    | O    | N    | S    |
|-------------------------|------|------|------|------|------|
|                         | 43.74| 4.56 | 28.21| 1.21 | 1.25 |

It can be seen from Table 1 that the content of volatile matter and moisture in Xiaolongtan lignite is very high and the content of fixed carbon is low, so the lignite has high activity and is easy to gasify. The results of element analysis showed that the content of carbon element in Xiaolongtan lignite was low, and the content of oxygen element and sulfur element was relatively high, so the content of sulfide was high in the combustion process and pneumatolysis of Xiaolongtan lignite was meaningful.

2.2 Preparation of acid washed coal

Raw coal sample (YN-row): Lignite from Xiaolongtan, Yunnan. Coal sample washed by HCl (YN-HCl): YN-row mixed with 36% of HCl solution in the ratio of 1:2 by volume, which reacted at room temperature and stir at 200r/min for 24 hours, then filtered. Washed the filter cake with deionized water until the washing solution was neutral. The filter cake was dried at 105 ℃ for 4 hours to obtain acid washed coal.

2.3 The main chemical reactions

1) The total reaction formula of coal pyrolysis [8,11]:

\[
\text{coal} \rightarrow CH_4 + \text{Gaseous hydrocarbon} + CO + CO_2 + H_2 + H_2O + \text{Coke}
\] (1)

2) Heterogeneous reaction
\[
C + H_2O \rightarrow CO + H_2 \quad \Delta H = 135.0 \text{kJ/mol} \quad (2)
\]
\[
C + CO_2 \rightarrow 2CO \quad \Delta H = 173.3 \text{kJ/mol} \quad (3)
\]
\[
C + 2H_2 \rightarrow CH_4 \quad \Delta H = -84.3 \text{kJ/mol} \quad (4)
\]

3) Homogeneous reaction
\[
CO + H_2O \rightarrow CO_2 + H_2 \quad \Delta H = -38.4 \text{kJ/mol} \quad (5)
\]
\[
CO + 3H_2 \rightarrow CH_4 + H_2O \quad \Delta H = -219.3 \text{kJ/mol} \quad (6)
\]

2.4 Experimental index
1) Gasification yield
The gasification yield mainly refers to the volume of syngas obtained by gasification of unit mass of raw materials in standard state. The gasification yield includes dry gas yield and wet gas yield. In this paper, wet gas yield is used.

\[
\text{GY} = \frac{Q_s}{m_c}
\]

Where GY is gasification yield, \(Q_s\) is volume of syngas in standard state, \(m_c\) is weight of test sample.

2) Carbon conversion
Carbon conversion is defined as the ratio of the sum of the molar mass of all carbon containing components in syngas to the molar mass of each carbon element added.

\[
x_c = \frac{\sum n_i}{n}
\]

Where \(x_c\) is carbon conversion, \(n_i\) is molar mass of the \(i\)th carbon containing component, \(n\) is molar mass of carbon in coal.

3) Gasification efficiency
The gasification efficiency is the ratio of syngas calorific value to coal calorific value.

\[
\text{GE} = \frac{\text{HHV}_s \times Q_s}{\text{HHV}_c \times m_c} \times 100
\]

Where GE is gasification efficiency, HHVs is calorific value per unit volume of syngas, HHVc is calorific value per unit mass of lignite.

3. Result analysis
3.1 Effect of temperature
In this section, YN-row and YN-HCl with particle size of 0.5-0.6mm were used as samples, and the effect of bed temperature on lignite gasification characteristics was investigated under the condition of feed water flow rate of 10g/min.

1) Effect on carbon conversion and gasification yield
It can be seen from Figure 1 that the reaction activity of Yunnan Xiaolongtan lignite used in this paper was relatively high. With the increase of bed temperature, the carbon conversion and gasification yield increased rapidly; among them, the carbon conversion and gasification yield increased fastest from 800 °C to 900 °C, while the growth rate decreased after 900 °C. Water gas reaction \(C + H_2O \rightarrow CO + H_2\) (reaction(1)) which was main reaction was endothermic reaction, with the increase of bed temperature, more carbon reacted with water vapor to produce CO and \(H_2\), and the carbon conversion rate also increased. In addition, when the coal particles reacted at high temperature, the heat was transferred from the outer layer to the inner layer of the coal particles, and the pyrolysis reaction was also carried out from the outside to the inside. Therefore, with the increase of bed temperature, the heat and mass transfer effects became more obvious which increased the pyrolysis rate and produced more volatile matter. The melting layer on the surface of the particles was no longer bound and the reaction rate also accelerated.
For acid washed lignite, the gas yield and carbon conversion increased with the increase of bed temperature, which was similar to that of raw coal. However, the average yield of acid washed lignite decreased by about 0.3L/g, and the carbon conversion rate increased by about 3%.

2) Effect on gas composition

It can be seen from Figure 2 that with the increase of bed temperature, the content of H₂ and CO increased, the content of CO₂ decreased, and the content of CH₄ also decreased, but it was very limited. In the process of steam gasification, the reaction of water gas C+H₂O→CO+H₂ (reaction (2)) was an endothermic reaction, with the increase of bed temperature, the reaction was enhanced, which made the content of CO and H₂ increase. The carbon monoxide shift reaction CO+H₂O→CO₂+H₂ (reaction (5)) was a reversible exothermic reaction, and the reverse reaction was enhanced with the increase of bed temperature, so the content of CO₂ and H₂ decreased and the content of CO increased. However, the effect of bed temperature on water gas reaction was greater than that on carbon monoxide shift reaction.

Fig.2 Effect of bed temperature on composition of syngas

![Graph showing effect of bed temperature on carbon conversion and gasification yield](image1)

![Graph showing effect of bed temperature on composition of syngas](image2)
The boudouar reaction \( \text{C} + \text{CO}_2 \rightarrow 2\text{CO} \) (reaction (3)) was a reversible endothermic reaction. With the increase of bed temperature, the reaction equilibrium shifted to the right, resulting in the decrease of \( \text{CO}_2 \) content and the increase of \( \text{CO} \) content. However, according to Fabrizio Scala et al. [8], it was found that in the high temperature steam gasification reaction, the \( \text{CO}_2 \) concentration was low, and the water gas reaction was dominant, which was two orders of magnitude stronger than the buduard reaction. In addition, the hydrogenation reaction \( \text{C} + 2\text{H}_2 \rightarrow \text{CH}_4 \) (reaction (4)) and methanation reaction \( \text{CO} + 3\text{H}_2 \rightarrow \text{CH}_4 + \text{H}_2\text{O} \) (reaction (6)) were exothermic reactions, with the increase of bed temperature, the content of \( \text{CH}_4 \) should decrease. However, within the scope of this study, the intensity of Methanation and hydrogenation reaction was very weak, and \( \text{CH}_4 \) mainly came from the pyrolysis process.

Compared with the original coal gasification, the \( \text{CO} \) content in acid pickling lignite gasification increased greatly, the content of \( \text{H}_2 \) and \( \text{CO}_2 \) reduced greatly, and the content of \( \text{CH}_4 \) reduced slightly. Because the minerals in coal mainly effected the carbon monoxide shift reaction \( \text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 \) (reaction (5))[9-10], after the minerals in coal were removed, the water gas reaction \( \text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2 \) (reaction (2)) was basically unchanged, while the carbon monoxide shift reaction was inhibited, resulting in the reduction of \( \text{H}_2 \) and \( \text{CO}_2 \) yield, and \( \text{CO} \) accumulation. At this temperature, \( \text{CH}_4 \) basically came from the cracking of volatile matter and tar, while the removal of minerals was not conducive to the cracking of volatile matter and tar, so the content of \( \text{CH}_4 \) decreased slightly.

3) Effect on gasification efficiency

It can be seen from Figure 3 that the gasification efficiency increased with the increase of bed temperature, which increased rapidly from 750 °C to 900 °C and the increasing rate decreased from 900 °C to 950 °C. The main reasons for the above rule were: with the increase of bed temperature, the content of \( \text{CH}_4 \) with higher calorific value decreased, but the water gas reaction \( \text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2 \) (reaction (2)) was significantly enhanced, and more carbon converted into gas, which made the total amount of syngas increase, especially the combustible components in the syngas increased significantly, resulting in the increase of the calorific value of syngas with the increase of bed temperature. At the same time, the calorific value of coal did not change because of the constant coal feed rate and the gasification efficiency increased with the increase of bed temperature.

Compared with the raw coal, the gasification efficiency of acid washed lignite decreased at different bed temperatures. The main reason was that the contents of \( \text{H}_2 \) and \( \text{CH}_4 \) in the three kinds of combustible gases decreased, only the content of \( \text{CO} \) increased. Moreover, in the gasification reaction (2) and (5), \( \text{H}_2 \) and \( \text{CO} \) reduced and increased in the same proportion, so the gasification efficiency decreased finally.

![Fig.3 Effect of bed temperature on gasification efficiency](image)

3.2 Effect on feed water flow

This section investigated the effect of different feed water flow on gasification characteristics under the condition of coal particle size of 0.5-0.6mm.

1) Effect on carbon conversion and gasification yield
It can be seen from Figure 4 and Figure 5 that with the increase of feed water flow, the carbon conversion and gasification yield increased significantly, especially when the feed water flow increased from 5g/min to 10g/min. This was because with the increase of feed water flow, the concentration of water vapor in the reactor increased, the reaction of \( \text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2 \) (reaction (2)) was enhanced, and more carbon in semi coke converted into gas products, so the carbon conversion and gasification yield increased. On the other hand, the increase of feed water flow also led to the excess of steam and increased the gas flow velocity, so that more pulverized coal can not react in time. It was taken out of the fluidized bed, thus reducing the carbon conversion and gasification yield. Based on the above reasons, the increase of carbon conversion and gasification yield slowed down when the feed water flow was from 10g/min to 15g/min.

Compared with raw coal, the change trend of acid washed lignite gasification was similar to that of raw coal. However, under the same feed water flow, the gasification yield of acid washed lignite decreased significantly, while the carbon conversion slightly increased. For example, when the temperature was 900 ℃ and the feed water flow was 10g/min, the gasification yield decreased from 1.5864L/g to 1.3842L/g, and the carbon conversion increased from 86.8% to 87.4%. The reason may be that the minerals in coal inhibit the carbon monoxide shift reaction, resulting in the overall reduction of syngas. After the removal of minerals, the pore diameter became larger and the pore path was more smooth, the coal char reaction was enhanced and the carbon conversion increased.

![Fig.4 Effect of steam flow rate on carbon conversion](image1)

![Fig.5 Effect of steam flow rate on gasification efficiency](image2)

2) Effect on gas composition

Figure 6 showed the effect of feed water flow on gas composition of lignite gasification under the condition of bed temperature of 900 ℃ and coal particle size of 0.5-0.6 mm. It can be seen from Figure 6 (a) that with the increase of feed water flow, CO content slightly increased, H₂ content increased greatly, especially when the feed water flow increased from 5g/min to 10g/min. In Figure 6 (b), with the increase of feed water flow, CO₂ content decreased, CH₄ content decreased less than 10%.

The reason for the above changes was: with the increase of feed water flow, the concentration of
steam in the reactor increased, which made the reaction \( C + H_2O \rightarrow CO + H_2 \) (reaction (2)) and reaction \( CO + H_2O \rightarrow CO_2 + H_2 \) (reaction (5)) increased, while reaction \( CO + 3H_2 \rightarrow CH_4 + H_2O \) (reaction (6)) weakened. And the increase of reaction (2) was greater than that of reaction (5), so the content of CO was on the rise. Both reactions (2) and (5) were in favor of \( H_2 \) generation. Therefore, with the increase of feed water flow, \( H_2 \) content increased significantly.

![Fig. 6](imageurl)

**Fig. 6** Effect of steam flow rate on composition of syngas

The increase of CO\(_2\) content was mainly due to the enhancement of reaction (5). The content of \( CH_4 \) decreased with the increase of feed water flow, because reaction (6) moved in a direction unfavorable to \( CH_4 \) generation.

With the increase of feed water flow, the contents of \( H_2 \), CO and CO\(_2\) in the acid washing coal gasification process increased in varying degrees, while the content of \( CH_4 \) decreased slightly. Compared with the original coal gasification process, the change trend of gas content was similar, but the generation amount reduced in different degrees. After the minerals are removed from coal, the activity of volatilization analysis and tar pyrolysis reaction decreased, and the amount of gas generated decreased accordingly. On the other hand, without the catalysis of minerals, the rate and degree of carbon monoxide shift reaction decreased significantly, resulting in the reduction of \( H_2 \) and CO\(_2\) production and a large amount of CO accumulation. In conclusion, minerals in coal have a great influence on the generation of syngas.

3) Effect on gasification efficiency

Table 2 showed the effect of feed water flow on gasification efficiency when bed temperature was 900 °C and coal particle size was 0.5-0.6 mm. It can be seen from table 2 that the lignite gasification efficiency increased with the increase of feed water flow, especially when it was from 5g/min to 10g/min. The reason was that with the increase of steam concentration in the reactor, more carbon in the char participated in the water gas reaction, and a large number of combustible gas (\( H_2 \) and CO) generated, meanwhile the calorific value of coal in the reactor was constant. Therefore, the gasification efficiency increased with the increase of feed water flow. From 10g/min to 15g/min, the gasification
efficiency almost did not increase, which was due to the excess of steam and had no obvious promoting effect on the reaction.

Compared with the raw coal gasification, the gasification efficiency of acid washed lignite decreased. This was mainly due to the lack of catalysis of minerals in acid washed lignite and the decrease of combustible gas content.

Table 2 Effect of steam flow rate on gasification efficiency

| Steam flow (g/min) | 5   | 10  | 15  |
|-------------------|-----|-----|-----|
| Row coal gasification efficiency (%) | 92.8 | 95.1 | 96.1 |
| Acid washing coal gasification efficiency (%) | 86.4 | 93.4 | 94.2 |

3.3 Effect on coal particle size

This section explored the effect of feed coal particle size on gasification law when feed water flow was 10g/min, in which the feed coal particle size was 0.3-0.38mm (fine particles), 0.5-0.6mm (medium particles), and 0.7-1.0mm (coarse particles).

1) Effect on carbon conversion and gasification yield

Figure 7 and Figure 8 showed the effect of coal particle size on carbon conversion and gasification yield. It can be seen from the figure that both carbon conversion and gasification yield decreased with the increase of coal particle size.

There were two main factors affecting carbon conversion and gasification yield: on the one hand, the smaller the coal particle size, the larger the heating and reaction area per unit volume, the easier the internal pyrolysis and the faster the gasification rate, so that the reaction of steam completed sufficiently and gasification carried out thoroughly, and tar was easier to be converted into small molecules and produced more gas. On the other hand, the ash content in coal decreased with the decrease of coal particle size [11], and the devolatilization of pulverized coal occurred in the grinding
process, resulting in the decrease of volatile content, while the volatile matter in coal and alkali metal and alkaline earth metal in ash had a positive catalytic effect on gasification [12-13]. Under the experimental conditions, the influence of the former was greater than that of the latter, resulting in the increase of carbon conversion and gasification yield with the decrease of coal particle size. In addition, compared with temperature and feed water flow, the effect of particle size change on carbon conversion and gasification yield was not significant.

Compared with the original coal gasification, the carbon conversion of acid washed lignite increased, while the gasification yield reduced slightly, and the difference between different particle sizes was basically the same. The main reason was that after the removal of minerals, gasification reaction was only affected by particle size, while raw coal gasification was affected not only by particle size, but also by the effect of minerals on gasification catalysis.

2) Effect on gas composition

It can be seen from Figure 9 that the CO content of raw coal decreased and the content of CH₄ and CO₂ increased with the increase of coal particle size, the H₂ content reached the maximum when the particle size of feed coal was 0.5 ~ 0.6mm.

With the decrease of coal particle size, the specific surface area (reaction area) of pulverized coal increased, so the smaller the particle size, the easier the reaction of \( C + H_2O \rightarrow CO + H_2 \) (reaction(2)) and \( C + CO_2 \rightarrow 2CO \) (reaction (3)), and CO and H₂ increased accordingly. On the other hand, the content of volatile matter and ash in coal decreased with the decrease of particle size[14]. Therefore, with the decrease of coal particle size, the content of H₂ and CO₂ decreased, while the content of CO increased.

In conclusion, under the experimental conditions of this paper, the CO content increased with the decrease of coal particle size; the order of H₂ content from large to small was: 0.5 ~ 0.6mm > 0.3 ~ 0.38mm > 0.7 ~ 1.0mm; the content of CO₂ and CH₄ increased with the increase of particle size, which was mainly due to the increase of volatile matter.

![Fig.9 Effect of particle size on composition of syngas](image)

Compared with raw coal gasification, there was no catalytic effect of minerals in coal. In the experimental range of this paper, there was no maximum value of H₂ content when the particle size of coal feed is 0.5 ~ 0.6 mm. In addition, the content of H₂, CO₂ and CH₄ in syngas decreased in different proportions, while the content of CO increased significantly.

3) Effect on gasification efficiency

This section explored the influence of coal particle size on gasification efficiency at 900 ℃ and feed water flow of 10g/min. The results were shown in Table 3.

| Feed coal particle size(mm) | 0.3~0.38 | 0.5~0.6 | 0.7~1.0 |
|----------------------------|----------|---------|---------|
| Raw coal gasification efficiency (%) | 97.1 | 97.8 | 93.8 |
| Acid washing coal gasification efficiency (%) | 93.9 | 91.8 | 88.9 |

It can be seen from table 3 that the gasification efficiency of raw coal gasification was the highest when the particle size of feed coal was 0.5 ~ 0.6 mm. It was the combustible gas content and the
calorific value of coal that affected the gasification efficiency. Due to the constant coal feeding amount in the experiment, the calorific value was basically unchanged; as mentioned earlier, with the increase of coal particle size, the CH₄ content increased and the CO content decreased, while the H₂ content reached the maximum when the coal particle size was 0.5 ~ 0.6 mm. Therefore, the gasification efficiency of coal with different particle sizes was 0.5 ~ 0.6 mm > 0.3 ~ 0.38 mm > 0.7 ~ 1.0 mm.

The acid washed lignite decreased with the increase of coal particle size. Compared with the raw coal gasification, although the overall change trend was similar, all decreased with the increase of coal particle size. But the raw coal gasification had the change of synthetic gas composition caused by the catalysis of minerals, which made the gasification efficiency maximum when the feed coal particle size was 0.5 ~ 0.6 mm. In addition, because of the influence of minerals on reaction (1), the gasification efficiency of acid washed lignite was lower than that of raw coal gasification.

4. Conclusion
The gasification characteristics of Yunnan Xiaolongtan lignite were investigated by using steam as gasifying agent on a fluidized bed test bed.

1) Bed temperature, feed water flow and coal particle size all had influence on lignite gasification in fluidized bed. With the increase of reaction temperature, the carbon conversion, gasification yield and gasification efficiency increased, while the contents of H₂ and CO in syngas increased and contents of CO₂ and CH₄ decreased. With the increase of feed water flow, the carbon conversion, gasification yield and gasification efficiency increased, and the contents of H₂, CO and CO₂ increased, only CH₄ content decreased slightly. Under the condition of other operation conditions unchanged, with the increase of particle size, the carbon conversion, gasification yield and gasification efficiency increased. With the increase of diameter, carbon conversion and gasification yield decreased, CO content decreased, CO₂ and CH₄ content increased. In the range of this experiment, H₂ content and gasification efficiency reached the maximum value when the particle size of coal feed was 0.5 ~ 0.6 mm.

2) Using acid washing to remove minerals from coal, the pyrolysis weight loss behavior of coal before and after acid pickling was studied. The results showed that the pyrolysis rate of coal after demineralization was faster and more thorough.

3) The steam gasification characteristics of acid washed lignite were investigated in the fluidized bed reactor. The results show that: for acid washed lignite, with the increase of bed temperature, gasification yield, gasification efficiency and carbon conversion increased, the content of H₂ and CO increased, while the content of CO₂ and CH₄ decreased in a certain proportion; With the increase of feed water flow, the gasification yield, gasification efficiency and carbon conversion rate increased, the contents of H₂, CO and CO₂ increased in varying degrees, while the content of CH₄ decreased slightly; With the increase of feed water flow, the gasification yield, gasification efficiency and carbon conversion increased, while the content of CH₄ decreased slightly. With the increase of diameter, the carbon conversion, gasification yield and gasification efficiency decreased, the contents of H₂ and CO decreased, and the contents of CO₂ and CH₄ increased.

4) Compared with raw coal gasification, it was found that the carbon conversion increased slightly and the gasification yield and gasification efficiency decreased slightly; the content of H₂ and CO₂ in syngas decreased significantly, and the CO production increased significantly, but CH₄ content did not change significantly; With the change of bed temperature, feed water flow and coal particle size, the carbon conversion, gasification yield, syngas composition and gasification efficiency changed. The trend was similar.

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