Bamboo is the vernacular or common term for members of a particular taxonomic group of large wood grasses (subfamily Bambusoideae, family Andropogoneae/Poaceae). Bamboo encompasses 1250 species within 75 genera most of which are relatively fast-growing, attaining stand maturity within five years, but flowering infrequently. Moso bamboo (Phyllostachys pubescens) is originally from China, where it occurs extensively (20,000 km$^2$ or 60% of total bamboo cover); introduced to Japan. Stands of this species attain 15–20 m in height and 10–20 cm in culm (stem). After 2–5 years from maturing, fungus begins to form on the outside of the culm, which eventually penetrate and overcome the culm. Around 5–8 years later, the fungal growths cause the culm to collapse and decay. This brief life means culms are ready for harvest and suitable for use within about three to seven years. This process is very important to keep the mass of carbon fixation by photosynthesis in the renewal bamboo stands (Nath et al., 2009). They also reported that the carbon storage efficiency is varied in bamboo species stands. Harvested bamboo culm is utilized as paper, textile, board, handicraft, and fuel in Asian countries and South America (Scurkock et al., 2000). It is in the national interests to find and study alternative energy resources. Bamboo can be considered as a potential energy feedstock because most of the bamboo stands were left without utilization, and utilization of bamboo developed the bamboo stands to sustainable stands.

Gasification of biomass (wood-based materials, agricultural residues, forestry waste etc.) is a promising technology that provides competitive means for producing chemicals and energy from renewable energy sources (Bridgwater, 2003). The gas mixture from the gasification process can be utilized in both conventional and advanced technologies for production of electrical energy, thermal power and automobile fuel giving a positive environmental contribution as biomass fuels are considered as a carbon neutral (Cetin et al., 2005, Umeki et al., 2012). The gas mixture resulting from the gasification process can also be used to produce syngas which has a direct application in all hydro-treating operations, ammonia production, and the synthesis of methanol, dimethyl ether and super-clean liquid fuels, such as Fisher-Tropsch oil (Kim et al., 2013). The demand for hydrogen is on the rise due to increase utilization of hydrogen as a new fuel. Currently, hydrogen gas is mostly produced by process as the catalytic reforming of natural gas, liquefied petrol gas, and naphtha with steam. Such a process, however, results a significant amount of carbon dioxide emission and thereby contributed to the global warming. For this reason, alternative methods of producing

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(Received February 20, 2014)

Summary

We proposed the two stage gasification: first stage is carbonization of biomass for production of char and second stage is steam gasification of char. This system can produce the hydrogen-rich water gas without any by-products. To evaluate bamboo as the second stage gasification resource, carbonization of moso bamboo and gasification of its char were carried out using the small laboratory plant. Carbonization of bamboo from 600 °C to 800 °C produced suitable chars as the source for steam gasification. The gasification of these bamboo chars produced clean hydrogen rich gases without any by-products. The wood gas from the carbonization had sufficient HHV available for fuels as the heating source in gasification. The gasification rate was increased remarkably with raising the gasification temperature. The lower gasification temperature induced water gas shift reaction, which leads to change the water gas composition. Raising the gasification temperature also increased specific HHV and decreased gas yield. Raising steam supply rate decreased specific HHV and CO gas yield whereas H$_2$ and CO$_2$ gas yields were increased. In these cases, water-gas shift reaction acted an important rule. The reaction rates of water gas reaction ($k_1$), water-gas shift reaction ($k_2$) and Boudouard reaction ($k_3$) are calculated and the values well explain the gas composition produced under various gasification conditions.

Key words: moso bamboo, char, hydrogen-rich gas, steam gasification
hydrogen gas are being studied including direct gasification of biomass which is a renewable source (Wang et al., 2008, Kawamoto and Kuromachi, 2009). A critical review of the relevant literatures reveals that steam is the most widely accepted as a gasification agent for production of hydrogen gas mainly due to quality of the product gas and high yield of hydrogen (Mermoud et al., 2006). Xiao et al. (2007) evaluated the effect of gasification temperature on gas composition from bamboo using a lab-scale fluidized bed. Kinetic study on bamboo pyrolysis was studied to understand, model, and control the decomposition process (Edward et al., 2008).

The progress in the field of hydrogen production from biomass has been largely hampered by technical issues associated with tar formation, which reduces the efficiency of gas production and interferes with the equipment operation (Sun et al., 2011). Formation of tar compounds generally renders the product gas mixture unsuitable for commercial application. Numerous investigations have been conducted on the elimination/destruction of tar compounds formed during biomass gasification (Rath and Staudinger, 2001, Devil et al., 2003, Li et al., 2004, Han and Kim, 2008). Practical possibilities of tar removal/destruction methods involve (with exclusion of in-bed methods and multi stage methods) high medium temperature downstream methods, usually based on catalytic tar cracking (Sulk et al. 2012). The other methods (like hot electrostatic precipitators and adsorption by various solvents at temperatures below 400 ℃) can be considered rather as auxiliary methods (Zwalt, 2009).

Gasification process generally consists of two primary steps: initial pyrolysis and subsequent char gasification. The latter is a late-determining step that may control the overall conversion process (Zhang et al., 2008). Gasification of biomass chars has been developed to gain a better understanding of the char reactivity and kinetic modeling using steam and carbon dioxide as gasification agents. (Ceratin et al., 2004, Klose and Wolki, 2005, Roberts and Harris, 2007, Nilsson et al., 2012). Several studies have been reported on the gasification of biomass char from different kinds of biomass, e.g. grapefruits skin (Montesions et al., 2002), Japanese cedar (Matsumoto et al., 2009), sunflower shell (Haykiri-Acma et al., 2006), beech wood (Wolfgang and Michael, 2005), slash pine (Fermoso et al., 2009) and bagasse (Chaudhari et al., 2003). Yan et al. (2010) tried to produce hydrogen rich gas by steam gasification of charcoal byproduct from fast pyrolysis liquefaction process. They concluded that both high gasification temperature and introduction of proper steam led to higher yield of dry gas and higher carbon conversion efficiency. Umeki et al. (2012) proposed that untreated char produced from high temperature steam gasification system can be extracted from the gasifier and combusted to generate high temperature steam.

We have been proposed a new gasification system without tar formation using chars as the gasification sources. The system design is consisted of two reactors separated as follows: the first reactor is a carbonization furnace for char production and the second reactor is char gasification part. Wood gas and char produced in the first reactor are divided, wood gas including tars is led to outside of the second reactor and char is led to inside of the second reactor. Wood gas can be used as the heat source of the second reactor for gasification of practical plants. The purpose of this study is how to produce the clean hydrogen gas from steam with char more effectively. Therefore, various chars were prepared from bamboo and gasified with steam under various gasification conditions, such as gasification temperature and steam supply rate.

**MATERIALS AND METHODS**

1. **Raw materials**

Moso bamboo (*Phyllostachys pubescens*) was harvested from a bamboo stand in the forest research of Niigata prefecture located Murakami city, in Japan. The bamboo was cut to length approximately 5 × 5 cm chip, and dried in air. Proximate and chemical analyses of the bamboo were performed according to JIS.

2. **Carbonization**

Preparation of bamboo char was performed by charging 300 g of bamboo chips into a stainless steel reactor with length of 500 mm, internal diameter of 80 mm. The reactor was heated using a cylindrical electric furnace, and the temperature was monitored using K-type thermocouples placed inside and outside reactor as shown in Fig. 1. The reactor was heated for 180 min to given temperature (400 ℃, 600 ℃, 800 ℃), and then held for 180 min. Produced char at each carbonization temperature were called Char400, Char600 and Char800 respectively in this paper. Each carbonization was repeated for 3 times to produce more reliable data. The yield results were calculated by a following formula.

Char yield (％)=100× dried char weight (g)/dried bamboo weight (g)

The wood gases were induced to a cyclone and collected in a gas bag behind a gas flow meter recording produced gas volumes. The major gases including H₂, CO, CH₄ and CO₂ were determined on a gas chromatograph (J-Scientelab GC-7000T) equipped with a thermal conductive detector (TCD) and packed column (2 m × 3.2 mm activated carbon) using...
argon as a carrier gas. Column oven temperature was 70 ℃ and Current mA of TCD was 90. Higher heating values (HHV) of wood gases were calculated using each gases HHV ($H_2$: 12.75 MJ/Nm$^3$, CO: 12.63 MJ/Nm$^3$, CH$_4$: 39.72 MJ/Nm$^3$).

The structural characteristics of Char800 were observed using a scanning electron microscope (SEM, JEOL JSM-5310LV) without conductive coating.

### 3. Gasification

Gasification of bamboo chars were carried out at 800 ℃, 900 ℃, and 1,000 ℃ using the steam supply rate from 0.05 g/min/g to 0.15 g/min/g of bamboo char as a gasifying agent. A fixed bed gasifier was placed inside of the electrical furnace and 30 g char are put into the gasifier. Prior to gasification, air in a gasifier and a cyclone were replaced with nitrogen. After 60 min from start of heating, the gasifier reached to given temperature and then steam was supplied into the gasifier by controlling a water pump. Produced gases were passed through a cyclone and a gas holder. A gas meter was set behind of the cyclone. Produced water gases were collected from the gas holder for measurement of the water gas compositions. The gas analyses were carried out in the same manner of the carbonization methods. This two stage gasification process is illustrated in Fig. 2.

Gasification rates were calculated initial gasification period from 0 min to 90 min in which the range of the gas production rate was linear.

The chemical reaction mechanism of char gasification with steam is considered to be very simple as described follows.

- Water gas reaction: $m_1 C + m_1 H_2O \rightarrow m_1 CO + m_1 H_2$ \hspace{1cm}$\cdots$(1) \hspace{1cm}$k_1$
- Water-gas shift reaction: $m_2 CO + m_2 H_2O \rightarrow m_2 CO_2 + m_2 H_2$ \hspace{1cm}$\cdots$(2) \hspace{1cm}$k_2$
- Boudouard reaction: $m_3 C + m_3 CO_2 \rightarrow 2 m_3 CO$ \hspace{1cm}$\cdots$(3) \hspace{1cm}$k_3$

Where, $m_1$ is reaction molar for water gas reaction.
$m_2$ is reaction molar for water-gas shift reaction.
$m_3$ is reaction molar for Boudouard reaction.

Then, weight of reacted carbon, and weight of produced $H_2$, $CO$ and $CO_2$ gas were calculated as follows.

- Weight of produced hydrogen (WH) = $2 (m_1 + m_2)$
- Weight of produced carbon mono oxide (WCM) = $28 (m_1 - m_2 + 2m_3)$
- Weight of produced carbon dioxide (WCD) = $44 (m_2 - m_3)$

Values of WH, WCM and WCD were obtained from the experimental results from gas composition and each gas volumes. Approximations of each reacted moles, $m_1$, $m_2$ and $m_3$ were derived from above equations. Reaction rate of $k_1$, $k_2$ and $k_3$ were calculated from $m_1$, $m_2$ and $m_3$ and the gasification rate.

### RESULTS AND DISCUSSION

#### 1. Proximate and chemical analysis

Table 1 depicts the proximate and chemical analysis of moso bamboo. Values of proximate analysis are typical of lignocellulosic materials. Generally, Volatile matter reflects to production yield of pyrolysates and fixed carbon reflects to char yield. Results of chemical analysis indicated the possibility of utilization of bamboo as biomass resource for production of char, water gas and bio-oil.

#### 2. Carbonization of bamboo

The results of carbonization of bamboo at various temperatures were listed in Table 2. Bamboo char yields were decreased with raising carbonization temperature. The char yield of carbonization at 400 ℃ was higher than that of the other carbonization at higher temperature. The gas from the char production process is called as wood gas, and can be used for fuel in the second stage process. Therefore, gas yield and specific HHV of wood gas are important factors for practical application of the wood gas as a fuel. The yield and the specific HHV of the wood gas produced from bamboo carbonization were increased with raising carbonization.

![Fig 2. Two stage gasification process.](image)
temperature, and the gas yields were intended to inversely proportional to the char yields. Carbonization at 400 °C yielded significantly low specific HHV caused by low CH₄ content in wood gas, as compared with higher carbonization temperatures. The specific HHV depended on the wood gas composition; CH₄ is particularly a higher calorific value than H₂ and CO. Generally, pyrolysis of biomass, such as gasification and carbonization, produces the tar as a by-product, which is decomposed to form hydrocarbon at high temperature pyrolysis (Li et al., 2004). It was considered that in the case of carbonization of bamboo at 400 °C, decomposition of the tar was not occurred and low CH₄ content of wood gas was yielded. Furthermore, CH₄ content of wood gas was a maximum value when carbonization temperature was 600 °C. This H₂ content was increased with raising carbonization temperature whereas the CO and CO₂ content was decreased with increasing carbonization temperature. These results indicated that carbonization was not carried out sufficiently less than at 600 °C.

Scanning electron micrographs of parenchyma tissue of a moso bamboo char (Char800) are shown in Fig. 3. Fig. 3-A is a SEM of transverse section of the bamboo char. Fig. 3-B is a magnified micrograph of a selected cell corner in a Fig. 3-A.

Table 2 Carbonization results of various temperature

<table>
<thead>
<tr>
<th>Carbonization results</th>
<th>Char400</th>
<th>Char600</th>
<th>Char800</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbonization time, min</td>
<td>360</td>
<td>360</td>
<td>360</td>
</tr>
<tr>
<td>Char yield, wt. %</td>
<td>35.0</td>
<td>30.4</td>
<td>25.8</td>
</tr>
</tbody>
</table>

Wood gas analysis

<table>
<thead>
<tr>
<th>Wood gas Volume, Nm³/kg char</th>
<th>0.21</th>
<th>0.31</th>
<th>0.39</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific HHV, MJ/Nm³</td>
<td>9.1</td>
<td>12.4</td>
<td>13.3</td>
</tr>
<tr>
<td>H₂</td>
<td>14.6</td>
<td>22.3</td>
<td>31.3</td>
</tr>
<tr>
<td>CO</td>
<td>41.3</td>
<td>26.1</td>
<td>26.1</td>
</tr>
<tr>
<td>CH₄</td>
<td>4.9</td>
<td>15.3</td>
<td>14.7</td>
</tr>
<tr>
<td>CO₂</td>
<td>39.3</td>
<td>36.3</td>
<td>27.9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Gas composition, v%</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂</td>
<td>59.3</td>
<td>59.4</td>
<td>60.9</td>
</tr>
<tr>
<td>CO</td>
<td>28.4</td>
<td>25.8</td>
<td>26.0</td>
</tr>
<tr>
<td>CH₄</td>
<td>1.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>CO₂</td>
<td>11.3</td>
<td>14.7</td>
<td>13.2</td>
</tr>
<tr>
<td>k₁, mol/min/kg of char</td>
<td>1.04</td>
<td>0.95</td>
<td>0.99</td>
</tr>
<tr>
<td>k₂, mol/min/kg of char</td>
<td>0.29</td>
<td>0.35</td>
<td>0.33</td>
</tr>
<tr>
<td>k₃, mol/min/kg of char</td>
<td>0.03</td>
<td>0.04</td>
<td>0.02</td>
</tr>
</tbody>
</table>

*Gasification temperature was 900 °C and steam supply rate was 0.05 g/min/g of char.

Fig 4. Water gas production curves from Char400, Char600 and Char800.

Table 3 Gasification results of bamboo char produced by various temperatures

<table>
<thead>
<tr>
<th>Gas properties</th>
<th>Char400</th>
<th>Char600</th>
<th>Char800</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas yield, Nm³/kg</td>
<td>4.93</td>
<td>4.95</td>
<td>4.89</td>
</tr>
<tr>
<td>Gasification rate, Ndm³/min/kg of char</td>
<td>63.1</td>
<td>58.9</td>
<td>58.8</td>
</tr>
<tr>
<td>Specific gas HHV, MJ/Nm³</td>
<td>11.7</td>
<td>11.0</td>
<td>10.8</td>
</tr>
<tr>
<td>Total gas HHV, MJ/kg of char</td>
<td>57.6</td>
<td>54.4</td>
<td>52.8</td>
</tr>
</tbody>
</table>

*Gasification temperature was 900 °C and steam supply rate was 0.05 g/min/g of char.
Smooth surface of cell wall is observed and it was shown that the cell wall structure of Char800 remained the same as that in the original bamboo cell structure.

3.1 Effect of char preparation temperature on water gas properties, gas composition and gasification rate.

Bamboo chars prepared various temperatures; Char400, Char600 and Char800 were gasified at 900 ℃ with steam supply of 0.05 g/min/g char. Internal and external temperature of a furnace are monitored during gasification period (per 15 min) as well as gas production volume. The results are shown in Fig. 4. The furnace temperature was moving up as a heating program. Water gas were produced after steam supplying, and the production rate were decreased around 120 min in each chars gasification. Gasification rates are calculated by linear zone of gas production curves from 75 min to 115 min. All of the gasification results were listed in Table 3. Char prepared at low temperature, such as Char400, is insufficient carbonization product included wood components without carbonization. These components are easily pyrolyzed to form tar-like materials as well as CH₄. Thus, gasification rate of Char400 was highest and produced water gas with a high specific HHV due to the high CO and CH₄ content in the water gas. Hydrogen content in the water gas from Char400 is highest over 60%. Water gas from Char600 and Char800 included hydrogen gas approximately 60%. Char preparation temperature did not effect on gas yield and hydrogen content in water gas.

In any case, the water gas reaction rate (k₁) is higher than the water gas shift reaction rate (k₂) and the Boudouard reaction rate (k₃), and greatly contributed to total gasification rate. k₂/k₁ values were 0.28 (Char400)–0.37 (Char600), and k₃/k₁ values were of 0.02–0.04. Therefore, k₃ can be ignorable for calculation of total reaction rate. There are no significant differences in these reaction rates among of various chars.

Water gas distributions were monitored by gas sampling per 30 min and analyzed gas components. The result of the case of Char800 is illustrated in Fig. 5 as production curves. Hydrogen gas content was increased as the gasification reaction proceeded. On the other hand, carbon monoxide was decreased as the reaction proceeded. In the case of carbon dioxide, significant difference was not observed during the reaction period.

3.2 Effect of gasification temperature on the gas yield, HHV and gas composition

The results of gasification of bamboo chars at various gasification temperatures were summarized in Table 4. Gasification temperature greatly influenced on the gasification rates and the gas distributions. The gasification rate was increased remarkably with raising the gasification temperature from 800 ℃ to 900 ℃. However, no significant effects on gas yield, specific HHV, and total gas HHV were founded with increasing the gasification temperature. A
comparison of gas compositions of water gas produced from various gasification temperatures is illustrated in Fig. 6. Carbon monoxide was increased and carbon dioxide was decreased with increasing gasification temperature. Hydrogen content in the water gas was not different within various gasification temperatures.

As mentioned above, the water gas reaction produce hydrogen and carbon monoxide, and the reaction rate is denoted $k_1$.

$$C + H_2O \rightarrow CO + H_2 \quad (k_1)$$

The water gas shift reaction produced carbon dioxide and hydrogen, and the reaction rate is denoted $k_2$.

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (k_2)$$

In this reaction, CO is supplied from water gas reaction. Thus, increasing the ratio of $k_1/k_2$ resulted in increasing CO content and decreasing CO$_2$ content. The ratio were 1.7 for 800 °C, 2.7 for 900 °C and 4.7 for 1000 °C, respectively.

Molar ratios of gas components of water gases from various gasification temperatures were calculated and these results were illustrated in Fig. 7. Ratio of H$_2$/CO from 800 °C gasification was higher than that from 900 °C and 1000 °C. In 800 °C gasification, the reaction rate of $k_2$ was approximately half of $k_1$. This result suggested that half of CO produced from water gas reaction 1 was consumed in gas shift reaction 2, and reacted with H$_2$O to form CO$_2$ and H$_2$.

3.3 Effect of steam supply on the gas yield, HHV and the gas composition

The results of gasification of Char800 at 900 °C in various steam supply rate from 0.05 g to 0.15 g/min/g char were listed in Table 5. Specific HHV and gas yield were decreased with raising the rate of steam supply, and H$_2$ and CO$_2$ yields were increased whereas CO yield was decreased as shown in Fig. 8. It was considered that the increase of steam concentration in a gasifier with raising steam supply rate accelerated water-gas shift reaction 2. Furthermore, raising the steam supply caused to decrease the gasification temperature. Yan et al. (2010) reported that raising steam flow rate decreased CO gas yield and increased H$_2$ and CO$_2$ gas yield in the gasification of char produced from fast
pyrolysis liquefaction. Thus, specific LHV was increased and total gas yield was decreased with the raising steam flow rate. Li et al. (2012) also indicated using municipal solid waste that CO gas yield was decreased and H\(_2\) and CO\(_2\) gas was increased with raising of mass ratio of steam to biomass. The effect of steam supply rate may result in the same tendency in most of biomass feedstock. Molar ratio of produced gas components are illustrated in Fig. 9. Ratio of H\(_2\)/CO was increased with increasing steam supply rate as well as H\(_2\)/(CO + CO\(_2\)) ratio. On the other hand, the ration of CO/CO\(_2\) was decreased with increasing the steam supply ratio. In the case of 1.5 ml/min gasification, the reaction rate of k2 was 37% of k1, and this means that 37% of H\(_2\) produced in the water gas reaction 1 was consumed in the water gas shift reaction 2. In the same manner, 46% and 48% of H\(_2\) produced in reaction 1 were consumed in reaction 2 for 3 ml/min and 4.5 ml/min, respectively. These consumption rates of H\(_2\) reflected to the ratios of H\(_2\)/CO.

### 3.4 Char gasification mechanism

In char gasification with steam, clean fuel gases were produced with high calories. The water gas composition and HHV were the result from the competing reactions within 1) Water gas reaction 2) Water-gas shift reaction 3) Boudouard reaction in various degrees.

The water gas reaction is a solid-gas heterogeneous reaction, and the reaction pathway should be more complex. Proposed gasification mechanism of char with steam for water gas reaction of 5C + 5H\(_2\)O → 5CO + 5H\(_2\) (first release of 4H radicals from char are not counted) was illustrated in Fig. 10. Surface structure model of char is expressed as a polycyclic polymer consisted of aromatic hydrocarbons (PAHs), as shown in Fig. 10. In first stage, char released H radicals from the char surface and acted as OH radical acceptor at the surface carbon radicals. Then –C–O–H structures are induced at the char surface, H radicals are released from –C–O–H again and the carbonyl structures are formed. Electron transfers occur at the adjacent C–C bonds and CO gas is formed. This chain reaction is continued on the char surface in the presence of H\(_2\)O. In this steam gasification, steam is diffused firstly in the lumen (intracellular space), and gasification occurred in the cell wall. Produced gases are released to the lumen and spread the outside of cell wall.

**REFERENCES**


竹炭の水蒸気ガス化による水素リッチガス生産

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（平成26年2月20日受理）

要　約

二段階ガス化は、1段目のバイオマスの炭化と2段目の炭化物の水蒸気ガス化より構成されている。このシステムは、副産物の生成を抑えて、水素リッチな水性ガスを生成することができる。2段目のガス化へのタケの利用を検討するために、モウソウダケの炭化とその炭化物のガス化を小型ラボプラントで行った。水蒸気ガス化に適した炭化物は、600 ℃から800 ℃で行われたタケの炭化にて生成された。これらの炭化物を用いたガス化では、副生成物がない水素リッチガスが生成された。炭化時に生成される木ガスは、ガス化の熱源の燃料として十分な高位発熱量（HHV）を有していた。ガス化温度は、ガス化温度の上昇と共に顕著に増加した。ガス化温度の低下は水性ガスシフト反応を誘導し、水性ガス組成の変化を生じる。また、ガス化温度の上昇は、生成ガス高圧発熱量を増加させ、ガス生成量を減少させた。水蒸気ガス化温度の上昇は、生成ガス高圧発熱量と、CO ガスの生成量を減少させたが、H2とCO2ガスの生成量を増加させた。これらの反応では、水性ガスシフト反応が重要な役割を果たす。計算によって求めた水性ガス反応 (A)、水性ガスシフト反応 (A2)、ブドウ酸反応 (A3) の反応速度は、様々なガス化条件下で生成されたガス組成によく合致している。

キーワード：モウソウダケ、炭化物、水素リッチガス、水蒸気ガス化

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