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論　文　内　容　要　旨

Study of improving and recycling of biomass by Soft-Hydrothermal-Process

Abstract

Recently, considerable effort has been directed to establishing a sustainable society that utilizes waste recycling, and a zero-emission non-CO2 gas production system. In order to establish a sustainable society, with practical material recycling, it is appropriate to focus on the character and application of Soft-Hydrothermal-Process.

The use of biomass is expected to spread as an energy resource to contribute to reduce the greenhouse gas emission because biomass does not increase the amount of carbon dioxide generated in the energy-using process. And biomass is also expected to be valuable resources for products, chemicals, food and feedstock.

Biomass is generally defined as "a large volume of material originated from plants to be renewable and biomass is evenly distributed in the world".

As Japan is a nation poor in resource, it imports about 80% of the total energy supply. In accordance with the agreement of COP3 (Third Conference of Parties to the U.N. Framework Convention on Climate Change) held in Dec. 1997, Japan has committed internationally to reduce greenhouse gas emission such as carbon dioxide, etc. by 6% of the 1990 level from 2008 to 2012.

Under such Circumstances, "Special Measures to Promote the Use of New Energy" was established in 1997 to positively introduce and promote the use of new energy from the viewpoints of stable securing energy and dealing with environmental problems, and in Jan. 2001 "biomass" was added as a new energy source.

The Biomass Nippon Strategy was decided by the Japanese government at a cabinet meeting held in December, 2002. It is Japan's first national strategy to utilize biomass as a valuable resource, and is comprehensive because it takes on viewpoints of the varied areas of technology, social and economic systems. This means that Japan has declared its decision to stop the devastation of resources. There are four basic reasons for adoption this strategy: Climate change prevention; Development of a recycling-oriented society; Incubation of new industries; and the activation of rural areas. Three types of goals have been set (technical, regional and national) with specific action plans for production, collection and transportation, conversion technologies, and stimulation of demand for energy or material uses. Through cooperation between several ministries, the implementation of the Strategy has been going on for the past one year, however there remain many challenges to be tackled including the high costs of using biomass and stimulating the demand for biomass products. Despite these hurdles, however, the utilization of biomass will continue to be an important policy issued in Japan.

The technologies for biomass utilization are grouped in two major areas: one is for energy and the other is for products.

Recently, certain technologies, to which the manufacture of raw materials for functional foods or chemical products from fishery waste or inedible portions of farming crops can be expected to emerge. For example, technologies of the extraction of DHA, EPA, γ-amino butyric acid, and dietary fiber, which are used as raw

materials for functional foods; chitosan, which is produced from crustaceans and used as raw material for anti-bacterial fiber, collagen, which is used as raw material for cosmetics; all have been turned to actual use. Development for the purposes of production of various substances, which can be utilized as raw materials equipped by diverse range of functionality in order to produce new medicines or materials that is already advanced. Thus, is far from the stage of verification, or is in the course of basic research.
Thus, a technology for disposal and recycling of waste biomass and unused biomass is desired. Soft-Hydrothermal-Process could be useful to accomplish this purpose. And Soft-Hydrothermal-Process is one of the advanced technologies for biomass and wastes, which have high moisture content, because it does not require a drying process.
Soft-Hydrothermal-Process is a high-enthalpy, low-density water vapor process, which has no coexisting liquids and, therefore, is removed from the boiling-point depth. relationship -- i.e. below saturated vapor pressure, but relatively high pressure. And Soft-Hydrothermal-Process has potential for promoting organic reactions, such as "selective separation" and "extraction" at 'so-called' soft hydrothermal conditions which can utilize the rejection heat.
But Soft-Hydrothermal-Process has been studied never, so a few is known about Soft-Hydrothermal-Process.
The objective of this study is to clarify the mechanism of "drying" and "extraction" in Soft-Hydrothermal-Process. Furthermore, improving and recycling waste biomass and unused biomass under Soft-Hydrothermal-Process was investigated. Laboratory animal bedding was chosen as a test material to represent refractory waste biomass. Harmful organic components were extracted from ordinary wood-chip bedding by Soft-Hydrothermal process, and the treated beddings were possible to use for maintaining of mice. On the other hand, Soft-Hydrothermal process decomposed and eliminated harmful organic components, excrement and ammonia from used wood-chip bedding. As a result, 85% of used beddings were possible to recycle. We also report the results of raising examination of mice using Soft-Hydrothermal process treated beddings. And micro-environment of room and cage was improved using wood-chip beddings treated by Soft-Hydrothermal process.

In Chapter 1, first of all, I have described the current status of biomass utilization in Japan, and the problem of global warming, which is one of the most critical environmental issues. So, biomass can be utilized as energy and as a basis for products, in the same way as fossil fuels, thus covering a wide range of applications across the entire range of daily human life. Consequently, the technology used for converting biomass into products, it is expected to put into practice the diversification of the products obtained through conversion, as well as endowing them with high added value. Besides, properties of Soft-Hydrothermal -Process with respect to drying and extraction have been discussed. Soft-Hydrothermal –Process technology could be useful to accomplish this purpose. Based on these descriptions, the aim and research method of this study have been presented.

In Chapter 2, in order to clarify mechanisms of drying and extract reaction of water for Soft-Hydrothermal-process, we examined drying characteristics by flow type reactor and extraction characteristics by batch type autoclave. Wood-chips, which are representative biomass material, were used in drying experiments. And several model organic compounds, which are derived from biomass, were used in extraction experiments. We can see from drying experiments that drying rate was increased at the increase of vapor density. It was suggested that monomolecular H2O of vapor phase draw out "bound water" from wood-substance due to monomolecular H2O weaken hydrogen bound of "bound water" and wood-substance, and then bind newly hydrogen bound with "bound water" . Besides, we can see from extraction experiments that the possibility of the selective extraction by controlling of reaction conditions, such as reaction time, pressure, and temperature of Soft-Hydrothermal-Process was investigated.

In Chapter 3, based on research above, we developed Soft-Hydrothermal-Process apparatus for improving and recycling the biomass. And we elucidated drying and extraction properties of the apparatus. On the base of experimental results, we proposed an optimum process to obtaining useful resources from the biomass by Soft-Hydrothermal-Process apparatus.

In Chapter 4, the feasibility of improving and recycling of waste biomass and unused biomass in Soft-Hydrothermal-Process was investigated by the apparatus as discussed in Chapter 3. Laboratory animal beddings were chosen as a test material to represent refractory waste biomass. Beddings for laboratory animals in their cages are an environmental factor that can influence on experimental data and well-being of animal, and contain volatile and harmful components, such as terpenes and aromatic compounds. In addition, a large amount of waste disposal beddings containing excrements are produced as an industrial waste from animal facilities of life

science institutes or breeding companies. Harmful organic components were extracted from ordinary wood-chip bedding by Soft-Hydrothermal-Process and were decomposed and eliminated harmful organic components, excrement and ammonia from waste disposal bedding by this process. Moreover, Soft-Hydrothermal-Process may improve harmful component-rich ordinary bedding and make waste disposal beddings possible to recycle for maintaining laboratory animals.

In Chapter 5, based on conclusions resulted from Chapter 4, we conducted raising examination of mice using this Soft-Hydrothermal process for treatment of beddings at the Tohoku University, Graduate School of Medicine, Institute for Animal Experimentation. Firstly, we evaluated the influence of Soft-Hydrothermal process on treatment of beddings by raise of mice. These examinations led to some conclusions as follows: (1) No significant differences existed in body weight (g) versus age of ICR mice, which were tested between ordinary bedding and Soft-Hydrothermal-Process treated beddings. (2) Similar results were found in data obtained at "Litter size of one female" and "Offspring weaned from female". The results indicated that there were not different influences, harmful effect and/or adverse effect to use Soft-Hydrothermal-Process treated beddings. And 85% of waste disposal bedding was possible to recycle as bedding. In other words, the Soft-Hydrothermal-Process treated bedding is utilized as ordinary bedding. Secondly, raising experiments of mice using Soft-Hydrothermal process treated beddings were carried out, and airborne dust particles and ammonia concentration were measured in an animal room and above mice cages. It was indicated that micro-environment of room and cages were improved using wood-chip bedding, which was treated by Soft-Hydrothermal process. Thus, Soft-Hydrothermal-Process treated beddings were not only to reuse again, but also their quality as bedding was improved distinctly.

In Chapter 6, the conclusions based on experiment data and results of discussions were presented. Our study clearly demonstrates that Soft-Hydrothermal-Process is a promising method for improving and recycling of biomass.

論文審査結果の要旨

本論文は、基礎的研究例がほとんどない乾燥水蒸気の特性を明らかにし、その具体的応用例として、実験動物の床敷に適用することにより、木質バイオマスの改質とリサイクル可能であることを示している。

第1 章では、未利用バイオマスと廃棄バイオマスの現状について述べ、既往のバイオマス利用技術とソフト水熱プロセス法の相違からソフト水熱プロセス法における乾燥水蒸気が木質バイオマスの改質とリサイクルに適する反応媒体であることを説明した。さらに、そのプロセスの応用として、動物実験施設等で大量に使用されている床敷を選定した理由について述べている。

第2 章では、ソフト水熱プロセス法での反応媒体としての乾燥水蒸気の特性を明らかにするため、木質バイオマスとして木質チップを用いて流通型の実験装置での乾燥特性と、木質バイオマス由来のモデル化合物を用いてオートクレーブでの抽出特性を実験を行っている。乾燥特性では、乾燥水蒸気の低密度で高エンタルピーの反応媒体であることが乾燥に有利なだけでなく、水蒸気が結合水を引き抜く乾燥水蒸気の乾燥メカニズムを推定した。抽出特性では水蒸気密度と反応温度および反応時間による選択的抽出特性を明らかにした。その結果、乾燥水蒸気は液相の高温高圧水にはない乾燥特性と液相の高温高圧水と同等あるいは同等以上の抽出特性を兼ね備えた反応媒体であること示している。

第3 章では、ソフト水熱プロセス法での反応媒体としての乾燥水蒸気の特性を利用した木質バイマスの実験装置を開発し、その性能特性について第2 章で明らかにした乾燥特性と抽出特性を考察した。実験装置の開発に際しては、ソフト水熱プロセス法での乾燥水蒸気を均質化する対策を講じて実用機レベルでの処理を可能にした。したがって、乾燥水蒸気の乾燥特性と抽出特性を兼ね備えた実用規模の実験機を開発できた。

第4 章では、第3 章で開発した実験装置により、木質バイオマスの改質と廃棄バイオマスの再生の可能性を実験動物の床敷を用い検討した。改質については、床敷としての機能を損なわないで木質チップの抽出成分に多く含まれている木質由来の化学物質を除去し、木質床敷の改質ができることを示した。また、再生については、床敷としての機能を損なわず、使用済み床敷材から腐敗物質を除去し、乾燥、滅菌・殺菌して再生することができることを示した。さらに再生床敷は、吸水性や脆性も優れ、粉塵量の減少による飼育環境の改善できることも示した。

第5 章では、ソフト水熱プロセス法により改質および再生した床敷についてマウス(slc：ICR♂、♀）による動物実験を行い、行動観察、体重曲線、産仔数、および離乳数より改質ならびに再生した床敷は実験動物に悪影響がないことを確認した。また、再生床敷による長期間飼育実験(33 週齢）の体重曲線と解剖検査による所見、摘出主要臓器の重量測定、血液学的検査(血色素量・赤血球数・白血球数・ヘマトクリット・MCV・MCH・MCHC)、および血液生化学的検査(総ビリルビン数、トリグリセライド、総コレステロール、グルコース、AST、ALt）による分析からも、再生した床敷が実験動物に悪影響がないことを確認した。したがって、従来のリサイクル技術では品質低下をもたらしたが、乾燥水蒸気処理を行った再生床敷は、床敷としての品質が向上していることを示した。

以上、本論文により、乾燥水蒸気領域での水の特性を利用して床敷の改質と再生が可能になったことにより、未利用バイオマスの改質と廃棄バイオマスのリサイクル、ならびに木質バイオマスの有効利用の可能性を明らかにした。このように、本論文は、２１世紀の環境科学に必要な少エネルギープロセスの一つである乾燥水蒸気利用技術の発展とその応用を示したもので、環境科学のみならず素材工学の発展に寄与するところが少なくない。

よって、本論文は博士（学術）の学位論文として合格と認める。