

## 学位論文内容の要旨

学位論文題目

A study on the application of water-film-forming-unit (WFFU) in enhancing carbon dioxide removal effectiveness using water absorption method

(水を吸収媒体に用いた二酸化炭素除去プロセスへの液膜形成装置 (WFFU) の導入によるその効率向上に関する研究)

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With increased global attention on the greenhouse effect and climate change, identifying an effective and economical solution to control the release of greenhouses gases, especially carbon dioxide (CO<sub>2</sub>), into the atmosphere has been the subject of much research. Because it does not use chemicals or produce toxic byproducts, water scrubbing is an environmentally friendly method of absorbing CO<sub>2</sub> from exhaust gas and therefore provides a promising means of controlling emissions of CO<sub>2</sub>. However, the great limitation of this method is a low interaction between CO<sub>2</sub> and water, resulting in a low degree of removal and a high-pressure (1.0 to 2.0 MPa) operating requirement. In this study, I employed an apparatus outfitted with one or several water-film-forming-units (WFFU) which can produce a large number of water-films along with fine bubbles to promote the mass transfer and contact between the gas and liquid phases and improve the effectiveness of water scrubbing.

The doctoral dissertation included 6 chapters and its content was presented as the following.

**Chapter 1** introduced the background, the objectives of this study and the structure of the doctoral dissertation.

The literature review related to this research and the summary of the previous study on the CO<sub>2</sub> removal technology were presented in **Chapter 2**.

In **Chapter 3**, the performance of an apparatus outfitted with a water-film generator in removing CO<sub>2</sub> from different concentrations of mixed gases (containing CO<sub>2</sub> and N<sub>2</sub>) while tap water as a physical solvent to absorb CO<sub>2</sub> was assessed through the obtained results of

removal efficiency and absorption rate under various conditions of key factors including internal pressure, gas supplying pressure, temperature, gas-to-liquid ratio (G/L), and initial CO<sub>2</sub> content. The internal pressure in the absorption tank and CO<sub>2</sub> initial content, have a significantly direct effect whereas temperature shows an inverse effect on the CO<sub>2</sub> removal efficiency and its absorption rate in water. The results also prove that the good performance of CO<sub>2</sub> removal process can be seen at the low gas supplying pressure of 0.30 MPa. The low value of G/L can increase the removal efficiency but it prevents the economic aspect due to a decrease of CO<sub>2</sub> absorption rate. On varying the experimental conditions – internal pressure (0.06 and 0.10 MPa), gas supplying pressure (0.30 – 0.71 MPa), temperature (10°C – 30°C), G/L (0.36 – 1.79), and initial CO<sub>2</sub> content (10% – 100%) – the CO<sub>2</sub> removal ability and absorption rate varied from 22.9% to 90.0% and  $4.5 \times 10^{-4}$  to  $44.4 \times 10^{-4}$  mol s<sup>-1</sup>L<sup>-1</sup>, respectively. For instance, the removal and absorption rates reached approximately 90.0% and  $12.0 \times 10^{-4}$  mol s<sup>-1</sup>L<sup>-1</sup>, respectively, when the experiment was operated at 10°C and 0.30 MPa of gas supplying pressure with 35% CO<sub>2</sub> inlet gas content and 0.71 G/L.

**Chapter 4** discussed about the application of statistical tools in assessing the performance of CO<sub>2</sub> removal process using the advanced water absorption apparatus. The influence of various parameters—pressure, initial CO<sub>2</sub> concentration, G/L, and temperature—on the CO<sub>2</sub> removal efficiency and its absorption rate in water were investigated and estimated thoroughly by statistical polynomial models obtained by the utilization of the response surface method (RSM) with a central composite design (CCD). Based on the analysis of experimental matrix containing 31 trials, a high efficiency of CO<sub>2</sub> capture can be reached in conditions such as low pressure, high CO<sub>2</sub> concentration at the inlet, low gas/liquid ratio, and low temperature. Furthermore, the coefficients of determination,  $R^2$ , were 0.996 for the removal rate and 0.982 for the absorption rate, implying that the predicted values computed by the constructed models correlate strongly and fit well with the experimental values. It evidences that the models can be used as useful tools to predict the CO<sub>2</sub> removal efficiency and absorption rate accurately without carrying out a large number of experiments. Therefore, the utilization of RSM-CCD can provide

several benefits such as time saving, reducing of experimental trials and availability for observing the interactions among factors.

As discussed above, the advanced apparatus equipped with one WFFU support for the CO<sub>2</sub> removal performance at low pressure but it still remains the limitation due to the low removal rate under high load of feed gas (low absorption rate at high G/L). So as to assess comprehensively the effect and the benefits of using WFFU in improving CO<sub>2</sub> removal process, I carried out the comparison of the values of CO<sub>2</sub> removal and absorption rate which obtained when conducting experiments in the apparatus equipped with non-, one- and two-WFFUs. The results and discussions for this matter was shown in **Chapter 5**. Based on our results, the WFFU significantly improves CO<sub>2</sub> capture at 0.30 MPa in a water absorption system with two WFFUs. The CO<sub>2</sub> removal rate was 20% greater than for conventional systems without WFFUs. Moreover, statistical data attained by the Taguchi analysis method showed that the number of WFFUs used in the absorption system has the greatest influence on CO<sub>2</sub> removal efficiency (contribution percentage = 50.65%) compared to gas supplying pressure, initial CO<sub>2</sub> concentration, G/L, and liquid temperature. I also thoroughly investigated the effects of these factors on CO<sub>2</sub> removal performance in the apparatus linked with non-, one- and two-WFFUs. The optimum conditions for CO<sub>2</sub> removal efficiency in a system equipped with two WFFUs are: low temperature, a G/L of 0.71, a gas supplying pressure of 0.30 MPa, and a high inlet CO<sub>2</sub> concentration. Therefore, our research improves on the physical absorption method for removing CO<sub>2</sub> from exhaust gas using tap water, thereby introducing a promising new technology for controlling carbon dioxide emissions in a more environmentally friendly manner.

Finally, **Chapter 6** summarized the findings of this research including the CO<sub>2</sub> removal performance when using WFFU in enhancing the water absorption process, the optimum removal conditions and the benefits of WFFU in the improvement of water absorption method. In this chapter, the suggestions for the further work was revealed.

# 学位論文審査の結果及び最終試験の結果報告書

## (博士後期課程博士用)

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【論文題目】			
A study on the application of water-film-forming-unit (WFFU) in enhancing carbon dioxide removal effectiveness using water absorption method (水を吸収媒体に用いた二酸化炭素除去プロセスへの液膜形成装置 (WFFU) の導入によるその効率向上に関する研究)			
【論文審査の結果及び最終試験の結果】			
<p>近年、温室効果ガスやそれによる気候変動などが世界的に注目される中、温室効果ガス、とりわけ二酸化炭素 (CO<sub>2</sub>) の効果的で低コストなコントロール手法の開発が望まれ、多くの研究がなされている。本研究では、多数の液膜 (水の泡) とファインバブルを同時に形成することで水中の気体交換効率を加速することが期待できる液膜形成装置を1つないし複数組み込んだ CO<sub>2</sub> 除去プロセスの開発を行った。</p> <p>本博士論文は6章で構成されており、その内容は以下の通りである。</p> <p>第1章では、本研究の背景と目的、本論文の構成について述べている。</p> <p>第2章では、CO<sub>2</sub> 除去技術に関する従来の研究についてまとめている。</p> <p>第3章では、液膜形成装置を組み込んだ CO<sub>2</sub> 除去プロセスの性能評価 (CO<sub>2</sub> 除去率及び CO<sub>2</sub> 吸収速度について) を行った。ガス供給圧力、CO<sub>2</sub> 含有率及び水温が CO<sub>2</sub> 除去効率に大きな影響を与えることが明らかとなり、ガス供給圧力が 0.3MPa の場合に高い CO<sub>2</sub> 除去効率を得られた。</p> <p>第4章では、統計的な手法 (RSM+CCD) を用いた解析から、ガス供給圧力は低めで、高い CO<sub>2</sub> 含有率、低い気液比の場合に高い CO<sub>2</sub> 除去率が得られることが明らかとなった。統計的な手法を用いて得られたモデル式による予測値は実測値をよく再現でき、このモデル式を用いて様々な予測を行えることが示された。</p> <p>第5章では、本プロセスの実用化を見据え、液膜形成装置の数を増加した場合の効果について検討した。実験では液膜形成装置の数を0, 1, 2個と変化させ CO<sub>2</sub> 除去効率に及ぼす影響を検討した。実験結果から液膜形成装置の数を増やすことで CO<sub>2</sub> 除去効率が大きく改善することが示された。さらに、統計学的解析 (TAGUCHI METHOD) の結果、ガス供給圧力、CO<sub>2</sub> 含有率、気液比、水温に比較して、液膜形成装置の数が CO<sub>2</sub> 除去効率と CO<sub>2</sub> 吸収速度に顕著に影響することが示された。液膜形成装置を2個設置した場合の最適運転条件は、水温は低く、気液比が0.7、ガス供給圧力が0.3MPa、高い CO<sub>2</sub> 含有率であることが明らかとなった。</p> <p>第6章では、以上をまとめて結論とし、今後の展望について述べている。</p> <p>公聴会には、学内外から42名の参加があり、活発な質疑応答がなされた。公聴会での主な質問内容は、①31通りの実験結果からモデル式の15のパラメータを推定しているが、このモデル式の精度を評価するときに31通りの実験結果を再現できるかどうかによって評価してよいのか(もともとフィッティングに用いた数値を再現できるのは当然ではないか)、②今後の課題で示した実験を実施して得られた結果をどのように研究に生かすのか、再度この博士論文で行っている統計的処理を行うのか、それともメカニズムを解明するために用いるのか、③本装置で生成する液膜のサイズはコントロールできるのか、できるとすれば CO<sub>2</sub> の溶解効率を向上できるのか、④装置内圧が変化している条件下 (装置ありと装置なし) で、直接 CO<sub>2</sub></p>			

溶解効率を比較できるのか、⑤何をもってCO<sub>2</sub>の溶解効率が向上したといているのか（単位体積あたりに溶解するCO<sub>2</sub>の量か、単位時間あたりに溶解するCO<sub>2</sub>の量か）、など多数であった。

以上のいずれの質問に対しても発表者からの的確で具体的な回答がなされた。

以上より、本研究は独創性、信頼性、有効性、実用性および完成度ともに非常に優れており、博士（工学）の学位論文に十分値するものと判断した。

論文内容および審査会、公聴会での質疑に対する応答などから、最終試験は合格とした。

なお、主要な関連論文の発表状況は下記の通りである（関連論文：3編）。

- 1) Diem-Mai Kim Nguyen, Tsuyoshi Imai, Thanh-Loc Thi Dang, Takaya Higuchi, Ariyo Kanno, Koichi Yamamoto and Masahiko Sekine, Response surface method for modeling the removal of carbon dioxide from a simulated gas using water absorption enhanced with a liquid-film-forming device, *Journal of Environmental Sciences*, Vol.65, pp.116–126, 2018.
- 2) Mai Kim Diem Nguyen, Tsuyoshi Imai, Wataru Yoshida, Loc Thi Thanh Dang, Takaya Higuchi, Ariyo Kanno, Koichi Yamamoto and Masahiko Sekine, Performance of a carbon dioxide removal process using a water scrubber with the aid of a water-film-forming apparatus, *Waste and Biomass Valorization*, 印刷中, 2018, DOI: <https://doi.org/10.1007/s12649-017-9951-8>
- 3) Diem-Mai Kim Nguyen, Tsuyoshi Imai, Shahira S. Aly, Takaya Higuchi, Ariyo Kanno, Koichi Yamamoto and Masahiko Sekine, Influence of water-film-forming-unit on the enhanced removal of carbon dioxide from mixed gas using water absorption apparatus, *Environmental Technology*, Accepted (10-Aug-2018).