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Brief Paper

# Microwave Chemistry as a Candidate of Electrification Technology toward Carbon Neutrality – Microwave Magnesium Smelting as an Example

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**SUMMARY** Japan encounters an urgent issue of "Carbon Neutrality" as a member of the international world and is required to make the action plans to accomplish this issue, i.e., the zero emission of $CO_2$ by 2050. Our world must change the industries to adapt to the electrification based on the renewable powers. Microwave chemistry is a candidate of electrification of industries for the carbon neutrality on the conditions of usage of renewable energy power generation. This brief paper shows an example of "Microwave Pidgeon process" for smelting magnesium in which heating with burning fossil coals can be replaced with microwave energy for discussing how microwave technology should be developed for that purpose from both the academic and industrial sides.
***key words:*** *Caron Neutrality, Electrification of industries, Energy saving, Microwave chemistry, Microwave heating, Internal heating, Rapid heating, Substance-selective heating*

## 1. Carbon Neutral Issues Encountered for Japan

A former prime minister, Suga, declared that Japan will accomplish the carbon neutrality at 2050 (see Fig.1). In addition, he stated at the Global Warming Countermeasures Promotion Headquarters and the Climate Summit held in USA in April 2021 that Japan will aim the reduction of $CO_2$ emission by 46% as a challenge matched with the 2050 target even to 50% as the further challenge [1]. Japan is the fifth country in the amount of $CO_2$ emission in the world, i.e., 11.1 Gt equivalent to 3.2% after China(32.1%), USA(13.6%), India(6.6%), Russia(4.9%) in 2020 [2]. The chemical industries are responsible for 20% in the whole industry $CO_2$ emission and the steel industries are so for 48%, clearly demonstrating the importance of the carbon neutrality in these industries in order to achieve the carbon neutrality targets declared by the Japanese government.

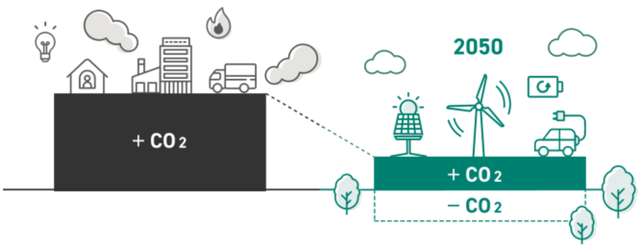


Fig.1 Concept by Ministry of the Environment

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## 2. Electrification of Industries as a Technology for Carbon Neutrality

A lot of research papers can be found in which the overview and future aspects are described on the basis of the investigations of worldwide initiatives, connotation, of carbon neutrality science, core technology, major challenges and strategies for carbon neutral, the position of new energy in the process of carbon neutral [3-7]. This short article would put a stress especially on microwave chemistry as a new technology enabling the carbon neutrality. Microwave heating was applied to a cooking oven in 1950s at the beginning of its history and has been applied to preparation technique of inorganic analysis, rubber vulcanization as an indirect heating technique as displayed in Fig.2. The two academic papers were published reporting the application of microwave heating to organic synthesis, stimulating chemists and chemical engineers to apply microwave techniques to synthesis of chemicals and materials in a lab-scale [8]. Meanwhile, researches on MW chemistry have been performed intensively not only by academics but also by industries, introducing the scope of scale-up in microwave technology even to large chemical plants [9-11]. Even chemical plants using solid catalysts are under the investigation towards the reform of the traditional chemical plants into a stage of electrification using microwave power.

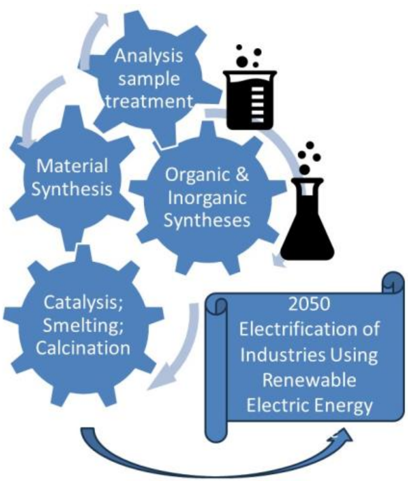


Fig.2 History of Microwave Chemistry

## 3. Magnesium Smelting by Microwave Power

### 3.1 Metal Treatments by Microwave Power

A lot of works can be found in which high-temperature treatments were attempted under microwave irradiation, exhibiting shortening of the treatment time, lowering the treatment temperatures for metal smelting and other processes [12-14]. Kashimura et al. proposed a design of microwave smelting furnace for iron ore with low $CO_2$ emission [13]. Now this article would take an example of magnesium smelting process, named "Microwave Pidgeon Method" for discussing the electrification of metal smelting process for carbon neutrality [15].

### 3.2 Microwave Pidgeon Method

Mg metal is produced by the Pidgeon process, in which dolomite as an ore is reduced with ferrosilicon at high temperature (1000-1300°C) under vacuum. This process requires a huge amount of energy consumption supplied by burning coal, resulting in the huge amount of $CO_2$ emission and release of PM2.5 particulates causing air pollution. The production of Mg (1 kg) requires burning of coal of 10.4 kg, emitting $CO_2$ of 37 kg.
Wada and Fujii made a challenge to replace burning of coal with microwave power of 2.45 GHz frequency aiming at electrification of the Pidgeon process [15]. Fig.3 shows a design of a microwave Pidgeon process reactor. The starting material molded as the briquettes consisting of the mixture of dolomite and ferrosilicon powders are placed in a ceramic-made container placed in a stainless steel-made applicator. The briquettes are irradiated by microwaves to heat up to the reaction temperature under vacuum. The produced Mg vapor is led to the collector and deposited on the wall of the collector.

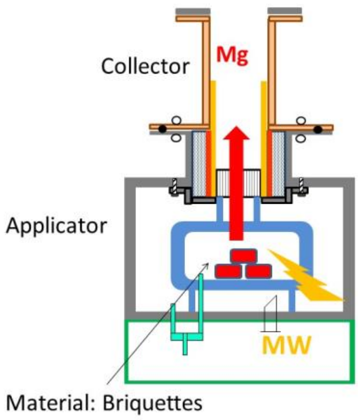


Fig.3 Design of Microwave Applicator

Fig. 4 shows photos of the actual reactor used in the lab, i.e., an overview photo of the applicator, an appearance of the heat-insulating ceramic container, a quarts tube containing the five briquettes in the vertical way. A block diagram in Fig. 4 shows a set-up of the microwave irradiation system, consisting of a magnetron, a tuner, a dummy load, a circulator, in infrared thermometer, a power meter, a waveguide.
They were successful in producing a mass of Mg metal (1.72 g) by 71.4% yield under the reaction conditions of 1000°C and 2 Pa, 80 min irradiation, 800W power as demonstrated in Fig. 5 as a photo of the produced mass and an XRD pattern observed for the product.

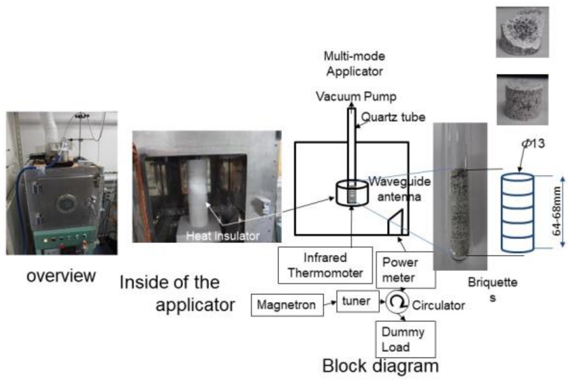


Fig.4 Lab Reactor Set-up of the MW Pidgeon Process

## 4. Estimation of Energy Consumption in the Microwave Pidgeon Process

Wada and Fuii's group attempted to design a scaled-up reactor and constructed a larger applicator shown in a photo in Fig.5 [15]. This large applicator was designed for producing over 10 g of Mg metal in a batch and investigated for the estimation of the energy consumption in the smelting process.

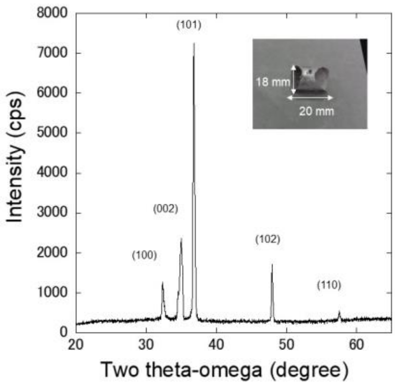


Fig. 5 Mg Metal Mass Produced by Lab MW-Pidgeon Process

Fig.6 shows a photo of an outer appearance of a large applicator. The block consisted of 108 briquettes (265 g) was heated up to 595 at the side wall of the block within 4.05 min, producing 7.12 g metal Mg (12% in the yield).
They estimated the energy consumption in this large applicator, taking into account the reaction heat in the reduction of dolomite with ferrosilicon, the heat capacity of the briquettes, the heating efficiency of the applicator and other components contained in and attached to the applicator, and the microwave generating efficiency of the magnetrons. The heating efficiency of the briquettes was estimated from the temperature rising behavior obtained in the experiments under microwaves to be 0.37 as the ratio on the basis of the whole energy input through the microwave energy input, in which the microwave energy input was estimated as the integrated value of the difference between the input power and reflectance power measured by the power meters attached to the applicator during the temperature rise expriments.

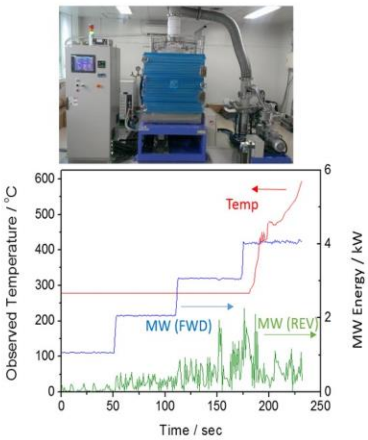


Fig.6 Large-scale Applicator and its Heating Behavior

Finally, the ratio of the energy consumption of the microwave large applicator (58.6 GJ/t-Mg) to that of the conventional Pidgeon process (192 GJ/t-Mg) was estimated as 0.31, meaning the energy saving by 69% in the MW Pidgeon process compared to the conventional Pidgeon process as the results of the above estimation.

## 5. Important Items for Discussion on Carbon Neutrality

The present example of MW Pidgeon process described in this paper teaches us how we can discuss properly about a new technology for electrification of a traditional and conventional process by replacing with a new technology. Magnesium is produced by smelting using a traditional Pidgeon process by combustion of coal causing huge $CO_2$ production. The authors of the MW Pidgeon process estimated the energy saving in the MW Pidgeon process by comparing with the traditional Pidgeon process. Fig. 7 displays a basic composition of a smelting furnace of the tradition Pidgeon process. A reactor called "retort" installed horizontally in the heating chamber is heated up to 1200℃ to induce the reduction reaction of dolomite in the briquettes with ferrosilicon by burning coal in the heating chamber on the right side and the produced Mg is deposited at the cooling part of the retort in the left side. In this traditional reaction set-up, not only the reaction part of the retort but also the other cooling parts are unavouldbly heated up, causing the unnecessary consumption of much energy. On the other hand, only the briquettes are heated up to 1200 ℃ in the MW Pidgeon method.

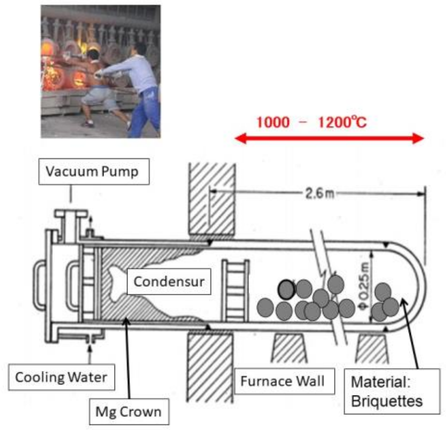


Fig.7 A Reactor of Traditional Pidgeon Process

Now I would address an important item for discussing advantages in using MW chemistry for electrification of industries on how the energy consumption is compared with conventional technology. This selective heating of the reaction field is the first important advantage in applying microwaves to the Mg smelting process instead of conventional coal burning. A reaction system is designed in an appropriate way depending on a new technology applied in a new process and is very different from a conventional system as demonstrated in the different decomposition of the reactors in Fig. 3 and 7.
As the second advantage in applying MWs for the smelting of magnesium in the Pidgeon process, the non-equilibrium local heating in the microscopic scale in the reaction field should be stressed here. Wada et al. demonstrated that the vibrating electric field is accumulated at the contact points of the particles constituting the reaction packing bed as shown in Fig. 8[17]. The reducing reaction of dolomite with ferrosilicon proceeds at the contact points between the dolomite and ferrosilicon particles heated higher than the other part by more than 100℃, resulting in an apparent lowering of the average reaction temperature. In other words, the energy necessary for the reaction is input selectively into the reaction field, the contact point of the reducing reaction in the MW Pidgeon process. This is the second advantage in applying MWs to the smelting technology.

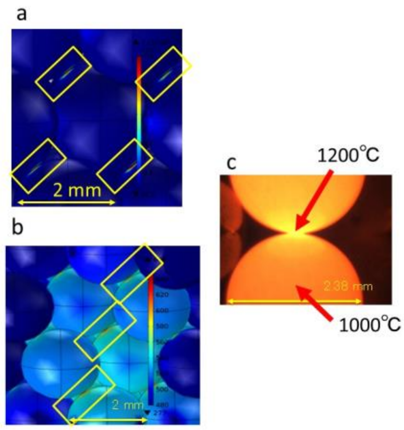


Fig.8 Local Heating at the Contact Points between Partcles in the Briquetts. a: E-field accumulated by 450 holds at the contact point between the particles predicted by the simulation with COMSOL Multiphysics. (shown as the bright lines at the contact points in the squares.) b: Local Heating at the Contact Points by the energy loss of Microwave Energy (higher by 100°C than the other part of the particle). c: Observed local heating at the contact point of the particle for SiC by the emission observation.

## 6. Summary

An example of electrification of a traditional energy-highly-consuming process by a new technology has been demonstrated, in which the energy saving is properly evaluated on the basis of the combined works of the lab experiments and the large scale reactor construction. Furthermore, a systematic study has revealed the mechanism of the temperature lowering of the reaction temperature, showing the reason for the energy saving. The author of the paper would emphasize the importance to perform and report the effectiveness of new green technologies in a proper and persuasive manner by showing both the scientific evaluations and experimental values in engineerings.

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### References

[1] Ministry of the Environment, https://ondankataisaku.env.go.jp/carbon_neutral/about/
[2] Japan Center for Climate Change Actions, https://www.jccca.org/about, https://www.jccca.org/download/66920
[3] L. Chen, G. Msigwa, M. Yang, A. I. Osman, S. Fawzy, D. W. Rooney, and P.-S. Yap, "Strategy to achieve carbon neutral society: a review," Environment Chemistry Letters, Vol.20, pp.2277-2310, 2022.
[4] Z. Caineng, X. Huaquig, X. Bo, Z. Guosheng, P. Songqi, J. Chengye, W. Ying, M. Feng, S. Qian, G. Chunxiao, and L. Minjie, "Connotation, innovation, and vision of "Carbon Neutrality"," Natural Gas Industry B, Vol.8, pp.523-537, 2021.
[5] Z. Caineng, X. Bo, X. Huaqing, Z. Dewen, G. Zhixin, W. Ying, J. Luyang, P. Songqi, and W. Songtao, "The role of new energy in carbon neutral," Petrol. Explor. Develop., 48(2), 480-491 (2021).
[6] T. Kochenburger, G. Liesche, J. Brinkmann, K. Gagalick, and D. Foertsch, "Fine chemicals production in a carbon-neutral economy: the role of electrification," Current Opinion in chemical Engineering, Vol.40, 100904, 2023.
[7] S. Ebrahimi, M. M. Kinnon, and J. Brouwer, "California end-use electrification impacts on carbon neutrality and clean air," Applied Energy, Vol.213, pp.435-449, 2018.
[8] A. de la Hoz, and A. Loupy, Microwaves in Organic Synthesis, Third, completely Revised and Enlarged Edition, Vol.1, Wiley-VCH Verlag & Co., Weinheim, Germany, 2012.
[9] Y. Kwak, C. Wang, C. A. Kavale, K. Yu, E. Selvam, R. Mallada, J. Santamaria, I. Julian, J. M. Catala-Civera, H. Goyal, W. Zheng, and D. G. Vlachos, "Microwave-assisted performace-advantaged electrification of propane dehydrogenation," Sci. Adv., Vol.9, eadi8219, 2023.
[10] I. A. Julian, and A. Fresneda-Cruz, "Chapter 17 Scale-up microwave-assisted heterogeneous catalytic processes," in Advances in Microwave-assisted Heterogeneous Catalysis, Catalysis Series, Royal Society of Chemistry, 2023.
[11] Y. Wada, S. Fujii, and S. Tsubaki, "Chapter Two Activation of chemical reactions on solid catalysts under microwave irradiation," in Advances in Green and Sustainable Chemistry, Microwaves; Ultrasounds; Photo- and Mechanochemistry and High Hydrostatic Pressure, Elsevier, 27-69 (2021).
[12] L. Xu, J. Peng, H. Bai, C. Srinivasakannan, L. Zhang, Q. Wu, Z. Han, S. Guo, and S. J. L. Yang, "Application of microwave melting for the recovery of Tin powder," Engineering, Vol.3, pp.423-427, 2017.
[13] K. Kashimura, K. Nagata, and M. Sato, "Concept of furnace for metal refining by microwave heating – A design of microwave smelting furnace with low CO2 emission," Materials Transactions, 51(10), 1847-1853 (2010).
[14] N. Mizuno, S. Kosai, and E. Yamase, "Microwave-based extractive metallurgy to obtain pure metals: A review," Cleaner Engineering and Technology, Vol.5, pp.100306, 2021.
[15] Y. Wada, S. Fujii, E. Suzuki, M. Maitani, S. Tsubaki, S. Chonan, M. Fukui, and N. Inazu, "Smelting magnesium metal using a microwave Pidgeon method," Scientific Reports, 7:46512, (2017).
[16] S. Ramakrisshnan, and P. Koltun, "Global warming impact of the magnesium produced in China using the Pidgeon p;rocess," Resources, Conservation and Recycling, Vol.42, pp.49-64, 2004.
[17] N. Haneishi, S. Tsuabaki, E. Abe, M. M. Maitani, E. Szuzuki, S. Fujii, J. Fukushima, H. Takizawa, and Y. Wada, "Enhancement of fixed-bed flow reactions under microwave irradiation by local heating at the vicinal contact points of catalyst particles," Scientific Reports, 9:222, 2019.

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