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研究課題名（和文） Understanding the three-dimensional multiscale porous microstructures by applying deep neural networks

研究課題名（英文） Understanding the three-dimensional multiscale porous microstructures by applying deep neural networks

研究代表者
Sciazko Anna（SCIAZKO, Anna）

東京大学・生産技術研究所・特任助教

研究者番号：３０８９８９４５

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研究成果の概要（和文）：機械学習を用いた微細構造解析フレームワークを開発した。本フレームワークの実装によって、解析精度の向上とともに観察・解析時間を短縮することができた。開発した解析フレームワークを用いて空隙率、コンポジット組成、粒子サイズなどを調整した固体酸化物形燃料電池（SOFC）のセリア系電極を定量的に評価した。このフレームワークを応用して、SOFC電極上に析出した炭素の構造を再構築・評価することに成功した。また、敵対的生成ネットワークを用いて2次元画像から人工的に3次元構造を作成する新しい手法も開発した。実構造データを用いて訓練することで、指定した特徴や組成・粒子分布勾配を有する構造の作成が可能となった。

研究成果の学術的意義や社会的意義
The porous media found interest in many fields of engineering. Particularly, the porous electrode microstructure determines the performances of fuel cells and batteries. This study proposed comprehensive framework for analyzing porous media microstructures based on machine learning methods.

研究成果の概要（英文）：An automated microstructure analysis framework using machine learning was developed. The framework was built to improve three-dimensional (3D) microscopy analysis by super resolution and semantic segmentation algorithms. The developed framework can contribute in shortening actual measurement time up to 8 times and data post-processing time by two orders of magnitude. Furthermore, a novel method with generative network was proposed to create an artificial 3D microstructure model from a single two-dimensional image. Additionally, the generative network trained with microstructure datasets, can fabricate realistic microstructure models with predefined properties and gradients. The automated segmentation framework was used to characterize gadolinium doped ceria ceria-based solid oxide fuel (SOFC) anodes with the controlled properties (porosity, material composition, and particle size) and it enables to reconstruct SOFC anode with carbon deposition.

研究分野： Energy engineering

キーワード： solid oxide fuel cell　machine learning　3D microstructure　FIB-SEM　GAN network　semantic segmentation　super-resolution　artificial structure

１．研究開始当初の背景

The **multiscale porous media** found interest in many fields of engineering. In particular, the porous electrode microstructure determines the performances of **fuel cells and batteries**. The multi-sized pore design is beneficial, because large pores enhance the gas transport and nano-pores increase active reaction area. In particular, the microstructure of anode determines **electrochemical performance of Solid Oxide Fuel Cell (SOFC)**. Although there are many studies discussing dependance between SOFC performance and their microstructural properties, the correlation is still not clear.

Most common method to analyze 3-D structures with feature size of 0.01 – 30 µm is **focus ion beam-scanning electron microscope** (FIB-SEM). However, the limitations of this technique for electrodes are attributed to the length scales of relevant microstructure features which cover several orders of magnitude. The rapid development of deep learning is revolutionizing many fields of engineering including material engineering, but its applicability to multi-scale porous media is limited. It is expected that **machine learning methods can help in characterization of SOFC microstructures** and finally help in **developing improved electrodes**.

２．研究の目的

(1) Providing **strategy to fabricate the porous electrode of SOFC**, considering balance between the diffusion management and electrochemically active reaction sites.
(2) Providing **new tools based on machine learning for microstructure studies in various fields of energy engineering and ceramics processing**. It aims to provide set of algorithms which enable quantitative microstructure-oriented studies on multi-sized porous materials. The approach by deep neural network can overcome FIB-SEM limitations and provide high resolution-large volume 3-D reconstruction.

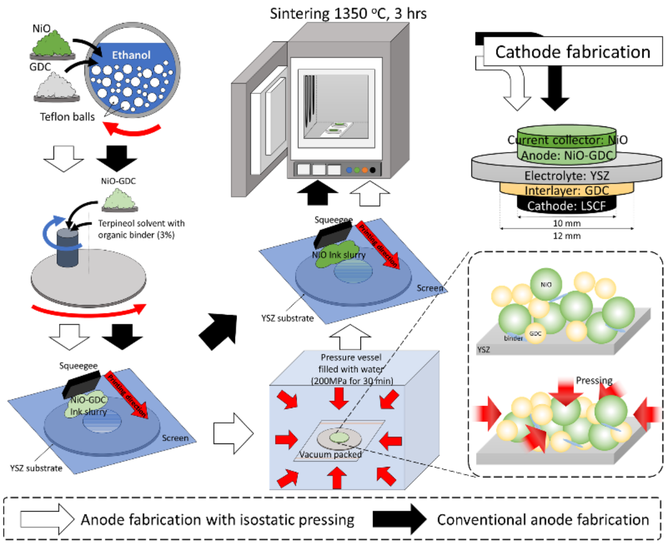

Fig.1 Fabrication of Ni-GDC electrodes with controlled properties. Komatsu, Y., Sciazko, A. and Shikazono, N., J. Power Sources, 485, 229317

３．研究の方法

(1) The SOFC samples with controlled particle size, porosity and phase fractions were fabricated (Fig. 1). The

fuel electrode was based on gadolinium doped ceria (GDC) with nickel or perovskite. The electrode performance was tested by series of electrochemical measurements including initial characterization and stability study. Samples are characterized by 2-D SEM and 3-D FIB-SEM.

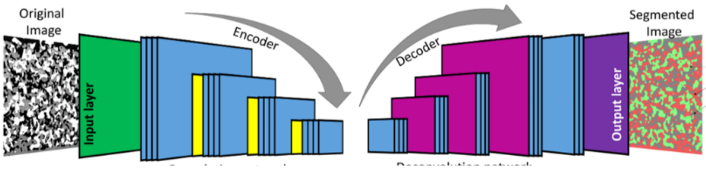

Fig. 2 Semantic segmentation network for SEM images of samples. Sciazko, A., Komatsu, Y., Shimura, T. and Shikazono, N., J. Electrochem. Soc., 168, 044504 (2021).

(2) The following algorithms for processing of microstructure data based on machine learning were developed:

- semantic segmentation algorithms for automatic processing large number of SEM images. The algorithm incorporates patch-based convolutional neural network (patch- CNN) in the encoder-decoder configuration (Fig. 2). The proposed network is utilized for segmentation of resin infiltrated cross-sectional images as well as data without resin infiltration.
- super-resolution algorithms for improving SEM and FIB imagining. The Very Deep Super Resolution (VDSR) network is applied to enhance FIB-SEM resolution in the cutting direction. The conditional generative adversarial network (C-GAN) is proposed for enhancing resolution of laser microscope image to SEM quality. (Fig. 3)
- algorithms for synthetic microstructure generation with 2-D and 3-D teaching data incorporating GAN^{2D-2D} and GAN^{3D-3D} networks. Additionally, C-GAN network is trained to fabricate the microstructures with

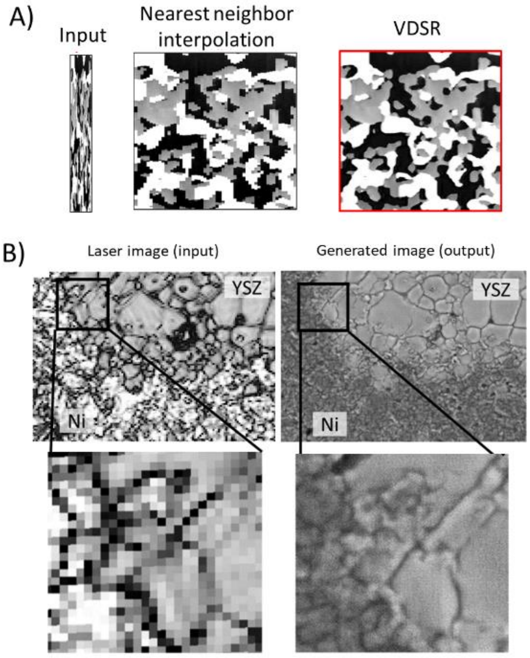

Fig. 3 Super resolution results: A) VDSR and B) C-GAN (Yamagishi, R., Sciazko, A., Ouyang, Z., Komatsu, Y., Nishimura, K. and Shikazono, N., ECS Trans., 103, 2087 (2021)).

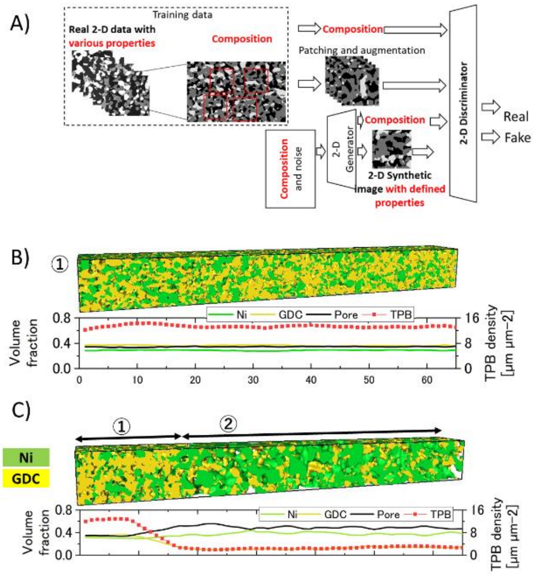

Fig. 4 A) C-GAN for generating microstructures with predefined properties B) uniform microstructure and C) structure with gradient. Yamagishi, R., Sciazko, A., Komatsu, Y., and Shikazono, N., Proc. 241th ECS meeting, I06-1083, (2022).

predefined properties as particle size, porosity, and ceramic phase composition (Fig. 4).

- algorithm for reconstructing artificial 3-D model from 2-D cross-sectional image introducing GAN^{2D-3D} networks. The proposed GAN network includes 3-D generator and 2-D discriminator (Fig. 5).

４．研究成果

In this project a framework for automated SOFC microstructure reconstruction was developed basing on the wide range of machine learning techniques. The proposed semantic segmentation network enabled automatic processing of large SEM image datasets with **accuracy over 98% and decreased processing time by two ranks of order from days to hours** (Fig. 2). The second tackled problem was improving the focus-ion beam scanning electron microscopy (FIB-SEM) measurements by super-resolving of in-depth direction by the patch-VDSR residual network (Fig. 3A). The application of the algorithm enables **decreasing FIB-SEM measurement time up to 8 times without loss of the resolution**. Additionally, the C-GAN based algorithm was developed for **improving quality of laser microscope images** (Fig 3B).

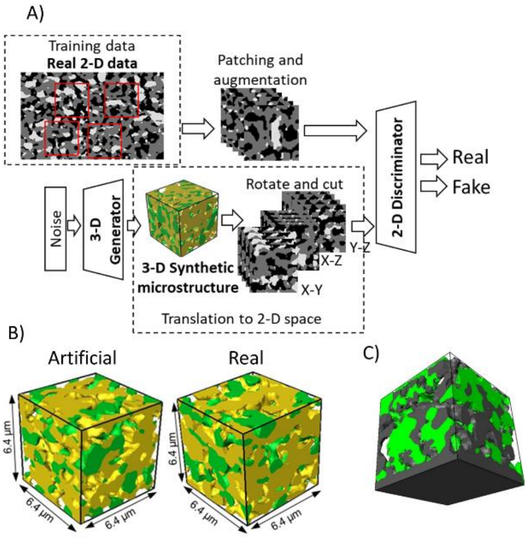

Fig.5 A) GAN2D-3D network for fabricating 3-D model from 2-D training data, generated structures B) isotropic microstructure and C) interface. Sciazko, A., Komatsu, Y. and Shikazono, N., ECS Trans., 103, 1363 (2021).

The GAN^{2D-3D} algorithm was proposed for **reconstructing artificial isotropic 3D model directly from 2D cross-sectional image** (Fig. 5A). The model is designed for the isotropic structures as shown in Fig. 5B. The weak GAN^{2D-3D} network was further developed for anisotropic materials enabling reconstructing microstructures with interfaces (Fig. 5C). Both visual quality and statistical characteristics of the artificial models are in very good agreement with real data. Moreover, C-GAN model was trained with microstructural database with various porosities, particle sizes and compositions as shown in Fig. 4A. The trained C-GAN network enables **fabricating realistic 2D and 3D artificial microstructure model with predefined properties and gradients** of properties along electrode (Fig. 4B-C).

The proposed algorithms were applied to automatic processing and analyzing data of GDC-based electrodes with various microstructural properties. The **GDC-based composites were prepared with nickel and perovskite materials and their performance was correlated with microstructures**. The examples of Ni-GDC anodes with various particle size, porosity and GDC share are shown in Fig. 6. The optimal condition for electrode fabrication was discussed in respect to the porosity, composition, and particle size. It was shown that the increased GDC composition, decreased porosity and decreased particle size resulted in the improved electrochemical performance as the number of electrochemical reaction sites increased (Fig. 7A). At the same time, the high GDC share over 80% in the composite may resulted in the porosity decrease and increase of gas diffusion resistance due to particle coarsening. Additionally, the degradation testing was conducted for Ni-based electrodes exposed to methane. **3D carbon structures in SOFC anodes are reconstructed for the first time** with the machine learning-assisted segmentation method without resin infiltration. It was shown that the nickel particles pulverize by metal dusting and ceramic support network is destroyed by the internal stress caused by carbon deposition. The correlation between electrode microstructure and carbon deposition was discussed highlighting influence of porosity and gas diffusion process (Fig. 7B).

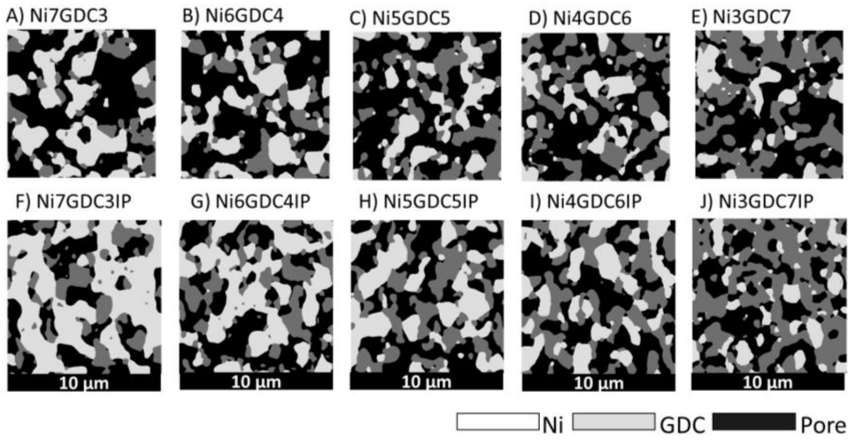

Fig.6 Microstructures of fabricated Ni-GDC samples with various porosities and Ni:GDC ratios.

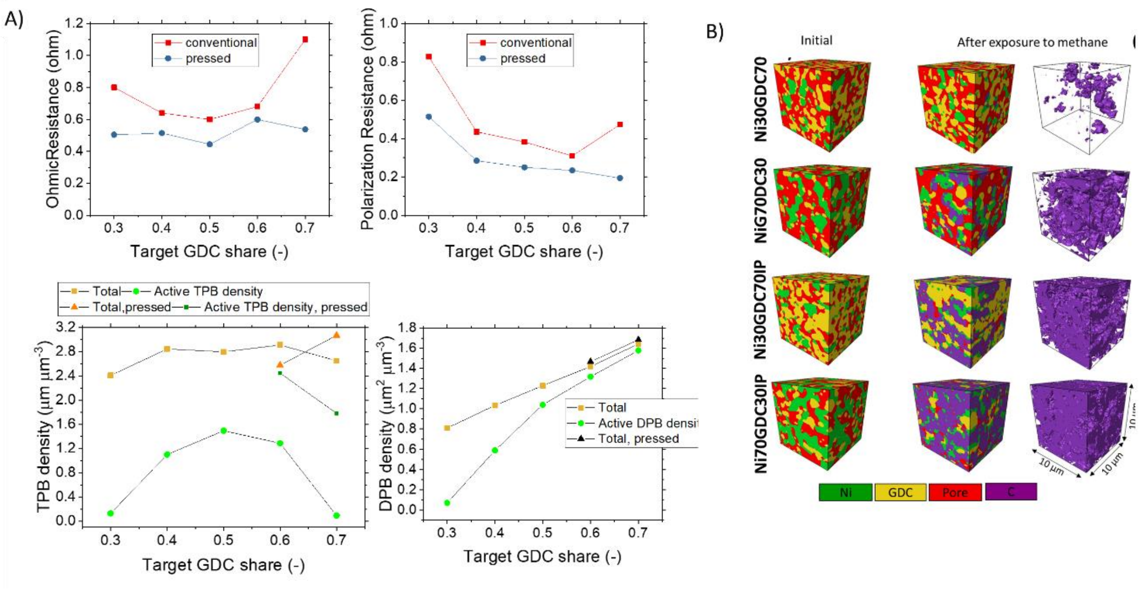

Fig. 7 A) Correlation between electrochemical performance and microstructures of Ni-GDC anodes B) results of the carbon deposition test for Ni-GDC anodes exposed to methane. Sciazko, A., Komatsu, Y., Nakamura, A., Sunada, Y., Ouyang, Z., Hara, T. and Shikazono, N., 15th European SOFC & SOE Forum, B1104, Lucerne, (2022).

５．主な発表論文等

〔雑誌論文〕　計3件（うち査読付論文　0件／うち国際共著　0件／うちオープンアクセス　0件）

１．著者名 Sciazko Anna、Komatsu Yosuke、Nakamura Akiko、Ouyang Zhufeng、Hara Toru、Shikazono Naoki	**４．巻** 460
２．論文標題 3D microstructures of solid oxide fuel cell Ni-YSZ anodes with carbon deposition	**５．発行年** 2023年
３．雑誌名 Chemical Engineering Journal	**６．最初と最後の頁** 141680～141680
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.cej.2023.141680	**査読の有無** 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	**国際共著** －

１．著者名 Sciazko Anna、Komatsu Yosuke、Yokoi Ryosuke、Shimura Takaaki、Shikazono Naoki	**４．巻** 42
２．論文標題 Effects of mass fraction of La0.9Sr0.1Cr0.5Mn0.5O3-δ and Gd0.1Ce0.9O2-δ composite anodes for nickel free solid oxide fuel cells	**５．発行年** 2022年
３．雑誌名 Journal of the European Ceramic Society	**６．最初と最後の頁** 1556～1567
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.jeurceramsoc.2021.11.039	**査読の有無** 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	**国際共著** －

１．著者名 Ouyang Zhufeng、Komatsu Yosuke、Sciazko Anna、Onishi Junya、Nishimura Katsuhiko、Shikazono Naoki	**４．巻** 529
２．論文標題 Operando observations of active three phase boundary of patterned nickel - Yttria stabilized zirconia electrode in solid oxide cell	**５．発行年** 2022年
３．雑誌名 Journal of Power Sources	**６．最初と最後の頁** 231228～231228
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.jpowsour.2022.231228	**査読の有無** 無
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２．発表標題
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46th International Conference and Expo on Advanced Ceramics and Composites (ICACC2022)（国際学会）

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３．学会等名
第26回動力・エネルギー技術シンポジウム講演論文集, 佐賀（招待講演）

４．発表年
2022年

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２．発表標題
Microstructures of Ni-GDC electrodes with carbon deposition

３．学会等名
15th European SOFC & SOE Forum, B1104, Lucerne, Switzerland（国際学会）

４．発表年
2022年

１．発表者名
Yamagishi, R., Sciazko, A., Komatsu, Y., and Shikazono, N.

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析出還元法によるナノNi-GDCアノードの発電特性と微細構造

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2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関