Supporting Information

Real-time imaging of the electric conductivity distribution inside a rechargeable battery cell

Seiju MATSUDA,[a,*,§] Shogo SUZUKI,[a] Kai YABUMOTO,[a] Hideaki OKADA,[a] Yuki MIMA,[b,c] Noriaki KIMURA,[b,c] and Kenjiro KIMURA,[a,c,d]

[a] *Graduate School of Science, Kobe University, 1-1, Rokkodai-cho, Nada-ku, Kobe-shi, Hyogo, 657-8501, Japan*

[b] *Integral Geometry Science, 1-5-6, Minatojima-minamimachi, Chuo-ku, Kobe-shi, Hyogo 650-0047, Japan*

[c] *New Energy and Industrial Technology Development Organization, 1310, Omiya-cho, Saiwai-ku, Kawasaki-shi, Kanagawa, 212-0014, Japan*

[d] *Center for Mathematical and Data Sciences, Kobe University, 1-1, Rokkodai-cho, Nada-ku, Kobe-shi, Hyogo, 657-8501, Japan*

* *Corresponding Author*: *staff-kimuralab@silver.kobe-u.ac.jp*

§ ECSJ Student Member

ORCID ID:

Seiju MATSUDA: 0000-0002-1637-9022

1. Basic equations of the current and the magnetic field in a non-existent space of the current

From Maxwell's equations, the static magnetic field satisfies the following equation in a space where there is no magnetic source.

$$\Delta H_i = 0 \quad (i = x, y) \qquad \cdots \qquad (1)$$

The general solution of this equation is expressed as the sum of the terms that increase exponentially in the z direction and the terms that decrease exponentially as follows.

$$H_i(x,y,z) = \frac{1}{(2\pi)^2} \iint e^{ik_x x + ik_y y} \left\{ a(k_x,k_y) e^{z\sqrt{k_x{}^2 + k_y{}^2}} + b(k_x,k_y) e^{-z\sqrt{k_x{}^2 + k_y{}^2}} \right\} dk_x dk_y \qquad \cdots \qquad (2)$$

k_x and k_y are wavenumbers in the x and y directions. $a(k_x, k_y)$ and $b(k_x, k_y)$ are functions represented by k_x and k_y. Let $z= 0$ be the z coordinate of the measurement surface. If the magnetic field source is only in the space where $z> 0$, $b(k_x, k_y)= 0$. Assigning the two-dimensional Fourier transform $f_i(k_x, k_y, 0)$ of the measured magnetic field distribution in Eq. (3) as the boundary conditions, the solution of Eq. (1) is derived by Eq. (4).[1]

$$f_i(k_x,k_y) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{-ik_x x - ik_y y} H_i(x,y,0)\, dxdy \quad (i = x, y) \qquad \cdots \qquad (3)$$

$$H_i(x,y,z_0) = \frac{1}{(2\pi)^2} \iint e^{ik_x x + ik_y y} \left\{ f_i(k_x,k_y) e^{z_0\sqrt{k_x{}^2 + k_y{}^2}} \right\} dk_x dk_y \qquad \cdots \qquad (4)$$

By substituting the magnetic field distribution $H_i\ (x, y, 0)$ obtained by the measurement into Eq. (3), (4), the magnetic field distribution $H_i\ (x, y, z_0)$ near the battery can be derived.

2. Analytical solution for current and magnetic field near the storage battery[2]

The target of this theory is a single-layer lithium-ion battery cell. The coordinate system has x, y axes along the conductor surface and the z axis in the normal direction. In Fig. S1, h_T is the distance between collectors; h is the thickness of the collector; z_0 is a coordinate of the electrode; σ_0 is the conductivity of the collector; $\sigma(x, y)$ is the two-dimensional conductivity distribution between the anode collector and the cathode collector in the battery; and $\varphi(x, y)$ is the two-dimensional potential difference distribution on the collector surface. δ is delta function, and δ' is a function obtained by differentiating the delta function in the z direction. The steady-state Maxwell equation is described as follows.

$$\nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t} = 0$$
$$\nabla \times \boldsymbol{H} = \boldsymbol{j} + \frac{\partial \boldsymbol{D}}{\partial t} = \boldsymbol{j} \quad \cdots \quad (5)$$

E and ***H*** are electric field and magnetic field, respectively. ***D*** and ***B*** are electric flux density and magnetic flux density, respectively. ***j*** is electric current. There is a potential φ in ***E***.

$$\boldsymbol{E} = -\nabla \varphi \quad \cdots \quad (6)$$

The x and y components of ***j*** are defined as follows using the in-plane current of the positive electrode $\boldsymbol{j}_{\mathbf{c}}$ and the in-plane current of the negative electrode $\boldsymbol{j}_{\mathbf{a}}$.

$$j_x = j_{x,\mathrm{c}} - j_{x,\mathrm{a}}$$
$$j_y = j_{y,\mathrm{c}} - j_{y,\mathrm{a}} \quad \cdots \quad (7)$$

The z component is defined as follows using σ and φ (φ_c is the potential distribution of the positive electrode current collector metal, and φ_a is the potential distribution of the negative electrode current collector metal.).

$$j_z = \frac{1}{h}\sigma\left(\varphi_\mathrm{c} - \varphi_\mathrm{a}\right) = \frac{1}{h}\sigma\varphi \quad \cdots \quad (8)$$

Since the in-plane conductivity $\sigma_{0,\,\mathrm{c}}$, $\sigma_{0,\,\mathrm{a}}$ of the positive electrode and the negative electrode is the conductivity of the metal, the non-uniformity of the spatial distribution and the difference between the positive electrode ($\sigma_{0,\,\mathrm{c}}$) and the negative electrode ($\sigma_{0,\,\mathrm{a}}$) are sufficiently small. Therefore, σ_0 is defined as follows.

$$\sigma_{0,\,\mathrm{c}} = \sigma_{0,\,\mathrm{a}} = \sigma_0 \quad \cdots \quad (9)$$

$\boldsymbol{j}_\mathrm{c}$ and $\boldsymbol{j}_\mathrm{a}$ are expressed as follows using the potential distributions φ_c, φ_a and $\sigma_{0,\,\mathrm{c}}$, $\sigma_{0,\,\mathrm{a}}$, respectively.

$$\begin{aligned} j_{x,\mathrm{c}} &= -\sigma_{0,\,\mathrm{c}} \frac{\partial}{\partial x}\varphi_\mathrm{c} \\ j_{y,\mathrm{c}} &= -\sigma_{0,\,\mathrm{c}} \frac{\partial}{\partial y}\varphi_\mathrm{c} \\ j_{x,\mathrm{a}} &= -\sigma_{0,\,\mathrm{a}} \frac{\partial}{\partial x}\varphi_\mathrm{a} \\ j_{y,\mathrm{a}} &= -\sigma_{0,\,\mathrm{a}} \frac{\partial}{\partial y}\varphi_\mathrm{a} \end{aligned} \qquad \cdot\cdot\cdot \qquad (10)$$

When Eq. (10) is used, Eq. (7) becomes Eq.(11).

$$\begin{aligned} j_x &= -\sigma_{0,\,\mathrm{c}} \frac{\partial}{\partial x}\varphi_\mathrm{c} + \sigma_{0,\,\mathrm{a}} \frac{\partial}{\partial x}\varphi_\mathrm{a} \\ j_y &= -\sigma_{0,\,\mathrm{c}} \frac{\partial}{\partial y}\varphi_\mathrm{c} + \sigma_{0,\,\mathrm{a}} \frac{\partial}{\partial y}\varphi_\mathrm{a} \end{aligned} \qquad \cdot\cdot\cdot \qquad (11)$$

From $\sigma_{0,\mathrm{c}} = \sigma_{0,\mathrm{a}} = \sigma_0$,

$$\begin{aligned} j_x &= -\sigma_0 \frac{\partial}{\partial x}\left(\varphi_\mathrm{c} - \varphi_\mathrm{a}\right) = -\sigma_0 \frac{\partial}{\partial x}\varphi \\ j_y &- \sigma_0 \frac{\partial}{\partial y}\left(\varphi_\mathrm{c} - \varphi_\mathrm{a}\right) = -\sigma_0 \frac{\partial}{\partial y}\varphi \end{aligned} \qquad \cdot\cdot\cdot \qquad (12)$$

From the above, the electric current on the electric current collector is described as follows.

$$\boldsymbol{j} = \sigma_0 \boldsymbol{E} = -\sigma_0 \nabla \varphi(x, y) \qquad \cdot\cdot\cdot \qquad (13)$$

From the second equation of Maxwell's equations, the magnetic field and current satisfy the following equations.

$$\begin{aligned} \nabla \times \boldsymbol{H} &= \boldsymbol{j} \\ \nabla \times \nabla \times \boldsymbol{H} &= \nabla \times \boldsymbol{j} \\ \nabla\left(\nabla \cdot \boldsymbol{H}\right) - \Delta \boldsymbol{H} &= \nabla \times \boldsymbol{j} \end{aligned} \qquad \cdot\cdot\cdot \qquad (14)$$

From the equation of Maxwell's equations,

$$\nabla \cdot \boldsymbol{B} = 0 \qquad \cdot\cdot\cdot \qquad (15)$$

If there is no magnetization in a homogeneous material, the magnetic permeability of the material is μ.

$$\begin{aligned} \nabla \cdot (\mu \boldsymbol{H}) &= 0 \\ \mu \nabla \cdot \boldsymbol{H} &= 0 \end{aligned} \quad \cdots \quad (16)$$

From the above, $\nabla \cdot \boldsymbol{H} = 0$. Substitute this into Eq. (14).

$$\Delta \boldsymbol{H} = -\nabla \times \boldsymbol{j} \quad \cdots \quad (17)$$

When this is applied to the conductor surface,

$$\Delta H_z = \nabla \times (\sigma_0 \nabla \varphi) = 0 \quad \cdots \quad (18)$$

With respect to the magnetic field distribution on the left side of this equation, there is no term related to the current $\boldsymbol{j}$ on the right side, which is the source term. Therefore, the magnetic field H_z does not depend on the current $\boldsymbol{j}$, so the z component of the magnetic field generated by the current is 0. Thus, the current flowing through the conductor electrode does not generate H_z. Assuming that the current is flowing in the z-axis direction inside the electrolyte, H_z will not be generated. This is because there are no defects such as via holes on the printed circuit board and current is generated uniformly.

Assuming that the thickness of the conductor electrode is h, the continuity equation of the current on the conductor substrate is as follows.

$$\left(\frac{\partial}{\partial x} j_x + \frac{\partial}{\partial y} j_y\right) h + j_z = 0 \quad \cdots \quad (19)$$

Rewriting the above equation using the two-dimensional conductivity distribution of the electrolyte gives the following.

$$-h \frac{\partial}{\partial x}\left(\sigma_0 \nabla_x \varphi\right) - h \frac{\partial}{\partial y}\left(\sigma_0 \nabla_y \varphi\right) + h_{\mathrm{T}}^{-1} \sigma(x, y) \varphi = 0 \quad \cdots \quad (20)$$

Rewriting this equation assuming that the conductivity of the current collector is constant is as follows.

$$-h\frac{\partial}{\partial x}\left(\sigma_0\nabla_x\varphi\right)-h\frac{\partial}{\partial y}\left(\sigma_0\nabla_y\varphi\right)+h_{\mathrm{T}}{}^{-1}\sigma(x,y)\varphi=0 \quad \cdots \quad (21)$$

The current flowing in the storage battery is expressed as follows.

$$\boldsymbol{j}=\left\{-\sigma_0\nabla_x\varphi(x,y),-\sigma_0\nabla_y\varphi(x,y),-h_{\mathrm{T}}{}^{-1}\sigma(x,y)\varphi(x,y)\right\}h\,\delta(z-z_0) \quad \cdots \quad (22)$$

The relationship between the magnetic field and the electric current is described as follows. The current flowing over the positive and negative electrodes is expressed as a gradient of φ. Therefore, all of these effects are taken into consideration in the following Poisson's equation with φ as the source term and the magnetic field as the left side.

$$\begin{aligned}
&\Delta H_x=h_{\mathrm{T}}{}^{-1}h\frac{\partial}{\partial y}\{\sigma(x,y)\varphi(x,y)\}\delta(z-z_0)-\sigma_0 h\left\{\frac{\partial}{\partial y}\varphi(x,y)\right\}\delta'(z-z_0)\\
&\Delta H_y=-h_{\mathrm{T}}{}^{-1}h\frac{\partial}{\partial x}\{\sigma(x,y)\varphi(x,y)\}\delta(z-z_0)+\sigma_0 h\left\{\frac{\partial}{\partial x}\varphi(x,y)\right\}\delta'(z-z_0)\\
&\Delta H_z=\frac{\partial}{\partial x}\left\{\sigma_0 h\frac{\partial}{\partial y}\varphi(x,y)\delta\left(z-z_0\right)\right\}-\frac{\partial}{\partial y}\left\{\sigma_0 h\frac{\partial}{\partial x}\varphi(x,y)\delta\left(z-z_0\right)\right\}=0\\
&\frac{\partial^2}{\partial x^2}\varphi+\frac{\partial^2}{\partial y^2}\varphi=(\sigma_0 h h_{\mathrm{T}})^{-1}\sigma(x,y)\varphi(x,y)
\end{aligned} \quad \cdots \quad (23)$$

The z coordinates of the magnetic field derived from the positive electrode and the magnetic field derived from the negative electrode differ by h_{T}, but this is small enough that it does not matter. In this equation, the z component is 0, but the others are not 0. Consider the x and y components. At this time, the following simultaneous equations can be considered.

$$\begin{aligned}
&\Delta H_x=h_{\mathrm{T}}{}^{-1}h\frac{\partial}{\partial y}\{\sigma(x,y)\varphi(x,y)\}\delta(z-z_0)-\sigma_0 h\left\{\frac{\partial}{\partial y}\varphi(x,y)\right\}\delta'(z-z_0)\\
&\Delta H_y=-h_{\mathrm{T}}{}^{-1}h\frac{\partial}{\partial x}\{\sigma(x,y)\varphi(x,y)\}\delta(z-z_0)+\sigma_0 h\left\{\frac{\partial}{\partial x}\varphi(x,y)\right\}\delta'(z-z_0)\\
&\frac{\partial^2}{\partial x^2}\varphi+\frac{\partial^2}{\partial y^2}\varphi=(\sigma_0 h h_{\mathrm{T}})^{-1}\sigma(x,y)\varphi(x,y)
\end{aligned} \quad \cdots \quad (24)$$

Substitute from the third expression into the first and second expressions.

$$\Delta H_x = h^2\sigma_0 \frac{\partial}{\partial y}\left\{\frac{\partial^2}{\partial x^2}\varphi + \frac{\partial^2}{\partial y^2}\varphi\right\}\delta(z-z_0) - \sigma_0 h\left\{\frac{\partial}{\partial y}\varphi(x,y)\right\}\delta'(z-z_0)$$
$$\Delta H_y = -h^2\sigma_0 \frac{\partial}{\partial x}\left\{\frac{\partial^2}{\partial x^2}\varphi + \frac{\partial^2}{\partial y^2}\varphi\right\}\delta(z-z_0) + \sigma_0 h\left\{\frac{\partial}{\partial x}\varphi(x,y)\right\}\delta'(z-z_0) \quad \cdots \quad (25)$$

$Q_x(k_x, k_y, z_0)$ and $Q_y(k_x, k_y, z_0)$ are assumed to be the two-dimensional Fourier transformation images of the x- and y-components, respectively, of the magnetic field, $H_x(x, y, z_0)$, $H_y(x, y, z_0)$.

$$\tilde{\varphi}(k_x,k_y) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{-ik_x x - ik_y y}\varphi(x,y)dxdy$$
$$Q_x(k_x,k_y,z_0) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{-ik_x x - ik_y y}H_x(x,y,z_0)dxdy \quad \cdots \quad (26)$$
$$Q_y(k_x,k_y,z_0) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{-ik_x x - ik_y y}H_y(x,y,z_0)dxdy$$

Using Eq. (26), Eq. (25) is rewritten as Eq. (27).

$$\frac{d^2}{dz^2}Q_x - ({k_x}^2 + {k_y}^2)Q_x = -h^2\sigma_0(ik_y)({k_x}^2 + {k_y}^2)\tilde{\varphi}\delta(z-z_0) - \sigma_0 h(ik_y)\tilde{\varphi}\delta'(z-z_0)$$
$$\frac{d^2}{dz^2}Q_y - ({k_x}^2 + {k_y}^2)Q_y = h^2\sigma_0(ik_x)({k_x}^2 + {k_y}^2)\tilde{\varphi}\delta(z-z_0) + \sigma_0 h(ik_x)\tilde{\varphi}\delta'(z-z_0) \quad \cdots \quad (27)$$

Then, we use the following Green function ($G_0(z, z_0, k)$).

$$G_0(z,z_0,k) = \frac{1}{2k}e^{-k|z-z_0|}$$
$$k = \sqrt{{k_x}^2 + {k_y}^2} \quad \cdots \quad (28)$$
$$\frac{\partial^2}{\partial z^2}G_0(z,z_0,k) - k^2 G_0(z,z_0,k) = \delta(z-z_0)$$

When Eq. (28)is used, Eq. (27) becomes Eq. (29).

$$Q_x(k_x,k_y,z) = \left\{-h^2\sigma_0(ik_y k^2)G_0(z,z_0,k) - \sigma_0 h(ik_y)\frac{d}{dz}G_0(z,z_0,k)\right\}\tilde{\varphi}(k_x,k_y)$$
$$Q_y(k_x,k_y,z) = \left\{h^2\sigma_0(ik_x k^2)G_0(z,z_0,k) + \sigma_0 h(ik_x)\frac{d}{dz}G_0(z,z_0,k)\right\}\tilde{\varphi}(k_x,k_y) \quad \cdots \quad (29)$$

In Eq., $z \rightarrow z_0$. The following equation is used for that purpose.

$$\lim_{z \to z_0+0} G_0(z, z_0, k) = \frac{1}{2k}$$
$$\lim_{z \to z_0+0} \frac{d}{dz} G_0(z, z_0, k) = -\frac{1}{2} \quad \cdots \quad (30)$$

The two-dimensional Fourier transform image of the magnetic field distribution is expressed as follows.

$$Q_x(k_x, k_y, z_0) = \frac{1}{2}\left\{-h^2\sigma_0(ik_y k) + \sigma_0 h(ik_y)\right\}\tilde{\varphi}(k_x, k_y)$$
$$Q_y(k_x, k_y, z_0) = \frac{1}{2}\left\{h^2\sigma_0(ik_x k) - \sigma_0 h(ik_x)\right\}\tilde{\varphi}(k_x, k_y) \quad \cdots \quad (31)$$

From Eq.(31), the equations for magnetic field and current are derived as follows.

$$ik_y Q_x(k_x, k_y, z_0) - ik_x Q_y(k_x, k_y, z_0) = \frac{1}{2} hk^2\sigma_0(hk-1)\tilde{\varphi}(k_x, k_y) \quad \cdots \quad (32)$$

The two-dimensional Fourier transform image of the electric potential is obtained as follows.

$$\tilde{\varphi}(k_x, k_y) = \frac{2\left\{ik_y Q_x(k_x, k_y, z_0) - ik_x Q_y(k_x, k_y, z_0)\right\}}{hk^2\sigma_0(hk-1)} \quad \cdots \quad (33)$$

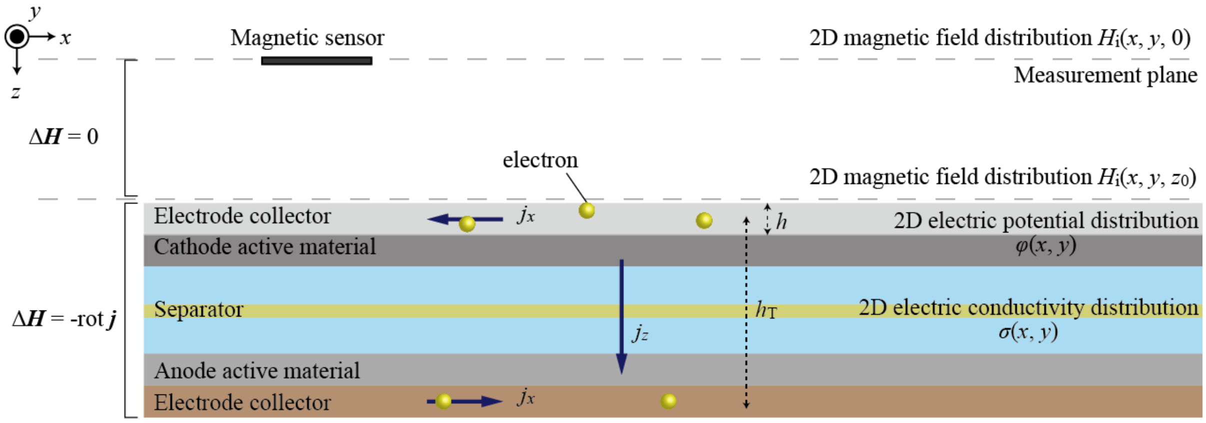
The two-dimensional conductivity distribution inside the storage battery can be obtained from φ obtained by the inverse Fourier transform.

$$\sigma(x, y) = hh_{\mathrm{T}}\sigma_0 \frac{\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\varphi}{\varphi} \quad \cdots \quad (34)$$

References

1. Y. Mima, N. Oyabu, T. Inao, N. Kimura and K. Kimura, Proceedings of IEEE CPMT Symposium Japan, **257** (2013).

2 K. Kimura, Y. Mima, and N. Kimura, *Subsurface Imaging Science & Technology*, **1**, 16 (2017).



S1. Definition of each variable of the process of reconstituting the electric conductivity distribution in the cell from the magnetic field leaking from the cell in the single-layer rechargeable battery.