Simple evaluation method of mechanical strength and mechanical fatigue of negative electrode for lithium-ion battery

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Abstract
Lithium-ion batteries (LIBs) are expected to be main power sources of automobiles. Nevertheless, LIBs easily lead to serious incidents because LIBs have high energy density. For application to automobiles, the reliability of LIBs should be guaranteed against various external loads. Especially, static loads and cyclic loads are constantly applied on LIBs because of vibration and thermal stress induced in automobiles, and fatigue damage occurs in electrodes of the LIBs. In this respect, it is important to evaluate mechanical strength and mechanical fatigue property of electrodes, such as tensile strength and S-N curves. This study has proposed a simple evaluation method of the mechanical strength and the fatigue property of electrodes for LIBs by using mechanical models of the electrodes. The actual alignment of particles of active material is random, and mechanical models based on the actual alignment are too complex to derive the main factor of mechanics of the electrodes. The proposed models approximate the alignment of the particles as the body-centered cubic (bcc) and the face-centered cubic (fcc) which are the well-known crystal lattices. In order to verify the proposed method, static tensile tests and bending fatigue tests of negative electrodes for LIBs have been conducted. From the test results, the tensile strength of the negative electrodes estimated by the proposed models agree with the experimental values, and the difference between the bcc model and the fcc model is smaller than the variation of the experimental values. The estimation value of the stress that initiates a crack on the negative electrodes by 1 cycle agrees with the tensile strength. The number of cycles linearly increases in the log scale with the decrease of the stress amplitude, and the stress amplitude at the $10^6$–$10^7$ cycles agrees with the half of the tensile strength.

Keywords: Battery, Electrode, Strength, Fatigue, Porous material, Thin film, Mechanical modeling

1. Introduction
Lithium-ion batteries (LIBs) are expected to be main power sources of automobiles. Nevertheless, LIBs easily lead to serious incidents because LIBs have high energy density. For application to automobiles, the reliability of LIBs should be guaranteed against various external loads. Especially, static loads and cyclic loads are constantly applied on LIBs because of vibration and thermal stress induced in automobiles, and fatigue damage occurs in electrodes of the LIBs. In this respect, it is important to evaluate mechanical strength and mechanical fatigue property of electrodes, such as tensile strength and S-N curves.

Over the past few years, many researchers have shown an interest in mechanical behavior of LIBs. Cai et al. (2011) and Ren et al. (2014) carried out macroscopic indentation tests on battery cells, and demonstrated change of voltage and temperature of the battery cells by internal short circuits. Rosso et al. (2006) reported initiation and propagation mechanisms of internal short circuits by microscopic observation. Dudézert et al. (2016) evaluated macroscopic electrochemical fatigue of LIBs subjected to cycles of charging and discharging by applying analogies between mechanical and electrochemical systems. Greve and Fehrenbach (2012), Sahraei et al. (2012a, 2012b, 2014, 2016), Lai et al. (2014), Ali
et al. (2015) and Zhang et al. (2017) have developed finite element simulations of battery cells. These studies presumed battery cells as composite structures of thin sheets; cover sheets, positive electrodes, negative electrodes and separator sheets. Using basic mechanical properties of the components of the battery cells, nominal mechanical properties of the battery cells were estimated. This approach is useful for simulating macroscopic behavior of LIBs in large machines such as automobiles.

On the other hand, the concern over microscopic mechanical behavior of LIBs has been risen. Electrodes of LIBs are active material (AM) coated on metallic foil. The AM on the metallic foil are formed into thin film. Hereinafter, this film is referred to as AM film in this study. Christensen and Newman (2004) has developed a mathematical model of electrochemical reaction of negative electrodes, and estimated thickness change of AM film during charging and discharging. Waldmann et al. (2014), Chen et al. (2019) and Uemura et al. (2019) have visualized mechanical behavior of AM film during charging and discharging in 5–200 μm scales by using X-ray computed tomography. Chen et al. (2018), Li et al. (2018), Yang et al. (2018) and Zhu et al. (2018a) have discussed crack initiation and propagation in AM film by external loads and electrochemical cycling based on scanning electron microscope (SEM) images. Most of the studies agree in that microscopic structure of AM film is bridged structure in which particles of AM are bonded by binder, and fracture of AM film is due to fracture of binder. There has been a great discussion about methodology to evaluate basic mechanical properties of AM film (Haselrieder et al., 2015; Lim et al., 2015; Westphal et al., 2015; Santimentaneedol et al., 2016; Giménez et al., 2018). In particular, numerical simulations by the discrete element method (Giménez et al., 2018) seems to be the most exact approach. However, actual alignment of particles of AM is random, and the high-cost simulations have to be iterated for enormous numbers of alignment cases.

This study has proposed a simple evaluation method of the mechanical strength and the fatigue property of electrodes for LIBs by using mechanical models of the electrodes. Because of the randomness of actual alignment of particles of AM, mechanical models based on the actual alignment are too complex to derive the main factor of mechanics of the AM films (Lai et al., 2014; Lim et al., 2015; Giménez et al., 2018; Zhu et al., 2018a, 2018b). The proposed models approximate the alignment of the particles as the body-centered cubic (bcc) and the face-centered cubic (fcc) which are the well-known crystal lattices. The binder is approximated to be elastic bars that joint the particles. In order to verify the proposed method, static tensile tests and bending fatigue tests of negative electrodes for LIBs have been conducted and discussed.

2. Manufacturing process of electrodes

Figure 1 shows macroscopic and microscopic structures of typical electrodes for LIBs. Electrodes for LIBs are AM coated on metallic foil. The AM of positive electrodes is Lithium transition metal oxide such as Lithium cobalt oxide (LiCoO2) and Lithium manganate (LiMn2O4). There are many kinds of chemical compounds for positive electrodes. In contrast, the AM of the negative electrodes is carbon-based material, and graphite is most-used. The metallic foil functions as current collectors. Copper foil and Aluminum foil are usually used because they have low electric resistance.

There are many manufacturing methods of electrodes in order to improve electrogenic capability. In these manufacturing methods, the common manufacturing process is as follows. First, powders of AM and binder are mixed with solvent. The solvent dissolves only the binder. The combination of the binder and the solvent is often polymer and organic solvent. Second, the slurry of the AM, the binder and the solvent is coated on metallic foil. The thickness of the coated slurry is controlled in order to achieve uniform thickness. Third, the coated slurry is dried by heating.

The macroscopic structure of the dried slurry is thin film (referred to as AM film). The microscopic structure of the AM film is bridged structure in which the particles of the AM are bonded by the binder. The binder supports the structure, and the stiffness and strength of the AM film is determined by the stiffness and strength of the binder. Electrochemical reactions for charging and discharging occur at pores in the AM film.

3. Mechanical modeling

Figure 2 shows schematics of macroscopic orientation and microscopic models in the proposed mechanical modeling of AM film. This figure shows the case in which the tensile direction is <100>. Actual alignment of particles of AM is random, and mechanical models based on the actual alignment are too complex to derive the main factor of mechanics of the AM film (Giménez et al., 2018). The proposed models approximate the alignment of the particles as the body–centered cubic (bcc) and the face-centered cubic (fcc) which are the well known crystal lattices.
Fig. 1 Structure of typical electrodes. In general manufacturing process, after powders of active material (AM) and binder are mixed with solvent, their slurry is coated on current corrector and dried by heating. Microscopic structure of AM film is bridged structure in which particles of AM are bonded by binder.

In Fig. 2, the spheres simulate particles of AM and the bars simulate binder. The particles of AM are assumed to be rigid sphere. The binder is assumed to be the elastic bars that joint the particles of AM and supports only the axial load. On this assumption, cross-sections along the binders seem to have the least strength. This paper focuses on the cases in which the tensile direction is vertical to the unit cube or the orientation direction of the binders; <100>, <110>, <112> for the bcc model and <100>, <110>, <111> for the fcc model.

3.1. Tensile direction <100>
If the tensile load $P$ is applied on the unit cube as shown in Fig. 2, the elongation $\lambda$ of the unit cube is presumed to be caused by the elongation of the binder. The nominal stress ($\sigma$) and the nominal strain ($\epsilon$) are described as follows.

$$\sigma = \frac{P}{L^2} \quad (1)$$
$$\epsilon = \frac{\lambda}{L} \quad (2)$$

where $L$ is the length of one side of the unit cube.

In actual measurements, only the composition ratios of the AM film can be measured. Using the volume ratio of the AM ($\alpha_a$) and the volume ratio of the binder ($\alpha_b$), the length ($l$) and the cross-section area ($A_t$) of the binder in the unit cube are derived by the following procedure. The volume of a particle of the AM ($V_a$) is

$$V_a = \frac{4\pi}{3} \left(\frac{d}{2}\right)^3 \quad (3)$$

where $d$ is the diameter of the particles of the AM. Then, the volume of the unit cube is described as follows.

$$V = \frac{nV_a}{\alpha_a} \quad (4)$$

where $n$ is the number of particles in the unit cube ($n = 2$ (bcc model), 4 (fcc model)). Therefore, the length of one side of the unit cube ($L$) is described as follows.

$$L = \sqrt[3]{V} = \left(\frac{nV_a}{\alpha_a}\right)^{1/3} = \frac{d}{2} \sqrt[3]{\frac{4\pi}{3\alpha_a}} \quad (5)$$

Then, using the middles of Fig. 2 (b) and (c), the length of the binder in the unit cube ($l$) is described as follows.

$$l = \frac{\eta L - 2d}{2} = \frac{L}{2} \left( \eta - \frac{2d}{L} \right) = \frac{L}{2} \left( \eta - 4 \sqrt[3]{\frac{3\alpha_a}{4\pi n}} \right) \quad (6)$$
Fig. 2  Macroscopic orientation and microscopic models. This figure shows the case in which the tensile direction is \( <100> \) as an example. Particles of AM are assumed to be rigid sphere and binder is assumed to be elastic bars. If the tensile load \( P \) is applied on the unit cube, the elongation \( \lambda \) of the unit cube is presumed to be caused by the elongation of the binder.

where \( \eta \) is constant (\( \eta = \sqrt{3} \) (bcc model), \( \sqrt{2} \) (fcc model) ). Likewise, the total volume of the binder \( (V_b) \) is

\[
V_b = \alpha_b V
\]  

(7)

and the cross-section area \( (A_t) \) of the binder is

\[
A_t = \frac{V_b}{N} = \frac{2\alpha_b L^2}{N} \left( \eta - 4\sqrt{\frac{3\alpha_a}{4\pi}} \right)^{-1}
\]  

(8)

where \( N \) is the coordination number (\( N = 8 \) (bcc model), 12 (fcc model)).

Next, the normal stress \( (\sigma_t) \) and the normal strain \( (\epsilon_t) \) of the binder are derived by the following procedure. Using the right of Fig. 2 (b), the normal stress \( (\sigma_t) \) and the elongation \( (\lambda_t) \) of the binder in the bcc model are described as follows.

\[
P = \frac{N \sigma_t A_t}{2} = \frac{4\sigma_t A_t}{\sqrt{3}} \quad \text{thus,} \quad \sigma_t = \frac{\sqrt{3}P}{4A_t}
\]  

(9)

\[
A_t = \left( \frac{L}{2} + \frac{\lambda}{2} \right)^2 + \left( \frac{L}{2} \right)^2 \approx \frac{L}{2} \left( \sqrt{3} + \frac{\lambda}{\sqrt{3}L} \right) = \frac{L}{2} \left( \sqrt{3} + \frac{2\lambda}{L} + \frac{\lambda^2}{L} \right) \approx \frac{L}{2} \left( \sqrt{3} + \frac{\lambda}{\sqrt{3}L} - \sqrt{3} \right)
\]  

(10)
Similarly, using the right of Fig. 2 (c), the normal stress ($\sigma_t$) and the elongation ($\lambda_t$) of the binder in the fcc model are described as follows.

$$P = \frac{N - 4^{\sigma_t}A_t}{2\sqrt{2}} = \frac{4\sigma_tA_t}{\sqrt{2}}$$

thus,

$$\sigma_t = \frac{\sqrt{2}P}{4A_t}$$

(11)

$$A_t = \sqrt{\left(\frac{L}{2} + \frac{\lambda}{2}\right)^2 + \left(\frac{L}{2}\right)^2} - \sqrt{\left(\frac{L}{2}\right)^2 + \left(\frac{L}{2}\right)^2} = \frac{L}{2} \left(\sqrt{22} + \frac{\lambda}{2\sqrt{2}L} - \sqrt{2}\right) = \frac{L}{2} \sqrt{2}$$

(12)

Equations (9)–(12) are summarized by using the constant ($\eta$) and Eqs (1), (2), (6), (8) as follows.

$$\lambda_t = \frac{1}{2\eta}$$

$$\lambda_t = \frac{\lambda}{2\eta} = \frac{1}{\eta L} \left(\eta - 4\sqrt{\frac{3\alpha_a}{4\pi}}\right)^{-1} = \frac{\epsilon}{\eta} \left(\eta - 4\sqrt{\frac{3\alpha_a}{4\pi}}\right)^{-1}$$

(15)

Hence, the equations between the macroscale (the nominal stress ($\sigma$) and the nominal strain ($\epsilon$) of the unit cube) and the microscale (the normal stress ($\sigma_t$) and the normal strain ($\epsilon_t$) of the binder) are described as follows.

$$\sigma = \frac{8\alpha_b}{\zeta N} \cdot \sigma_t$$

(16)

$$\epsilon = \zeta \epsilon_t$$

(17)

where

$$\zeta = \eta \left(\eta - 4\sqrt{\frac{3\alpha_a}{4\pi}}\right)$$

(18)

Equation (16) means that the nominal strength of the AM film ($\sigma$) is proportional to the volume ratio of the binder ($\alpha_b$). The AM film with the rich binder will achieve high strength.

### 3.2. Tensile direction \(<110>\)

Figure 3 shows the macroscopic orientation and the microscopic models in the tensile direction \(<110>\). In the bcc model, the total tensile load of the unit cuboid ($P$) is supported by four binders and described as follows.

$$P = \sigma_t A_t \sqrt{\frac{7}{3} \times 4} = \frac{4\sqrt{2} \sigma_t A_t}{\sqrt{3}} = \frac{4\sqrt{2} \sigma_t A_t}{\eta}$$

(19)

By using Eq. (8), the nominal stress ($\sigma$) is

$$\sigma = \frac{P}{\sqrt{2}L^2} = \frac{1}{\sqrt{2}L^2} \cdot \frac{4\sqrt{2} \sigma_t A_t}{\eta} = \frac{4\sigma_t A_t}{\eta L^2} = \frac{4\sigma_t}{\eta L^2} \cdot \frac{2\alpha_b L^2}{N} \left(\eta - 4\sqrt{\frac{3\alpha_a}{4\pi}}\right)^{-1} = \frac{8\alpha_b}{\zeta N} \cdot \sigma_t$$

(20)

This equation is same as Eq. (16) that is the case of the tensile direction \(<100>\). The nominal strain ($\epsilon$) is derived as follows. By using the elongation of the unit cuboid ($\lambda$), the elongation of each binder ($\lambda_t$) is

$$\lambda_t = \sqrt{\left(\frac{\sqrt{2}L}{2} + \frac{\lambda}{2}\right)^2 + \left(\frac{L}{2}\right)^2} - \sqrt{\left(\frac{L}{2}\right)^2 + \left(\frac{L}{2}\right)^2} = \frac{L}{2} \left(\sqrt{22} + \frac{\lambda}{2\sqrt{2}L} - \sqrt{2}\right) = \frac{L}{2} \sqrt{2}$$

(21)

Thus, by using Eq. (6),

$$\lambda = \sqrt{2\eta} \lambda_t = \sqrt{2\eta} \epsilon_t$$

(22)

$$\epsilon = \frac{\lambda}{\sqrt{2}L} = \frac{\sqrt{2\eta} \epsilon_t}{\sqrt{2}L} = \eta \cdot \frac{L}{2} \left(\eta - 4\sqrt{\frac{3\alpha_a}{4\pi}}\right) \cdot \epsilon_t = \frac{1}{2} \cdot \zeta \epsilon_t$$

(23)
In the fcc model, the elongation of the binder 1 ($\lambda_{1t}$) is equal to the elongation of the unit cuboid ($\lambda$). On the other hand, the elongation of the binder 2 ($\lambda_{2t}$) is

$$\lambda_{2t} = \sqrt{\left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2}\right)^2} = \frac{L}{2\sqrt{2}} - \sqrt{\left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2}\right)^2} \approx \frac{L}{2\sqrt{2}} - \sqrt{\left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2\sqrt{2}}\right)^2 + \left(\frac{L}{2}\right)^2} = \frac{L}{2\sqrt{2}} - \frac{L}{2} + \frac{L}{2} = \frac{L}{4} \tag{24}$$

This equation means that the normal stress ($\sigma_{2t}$) and the normal strain ($\epsilon_{2t}$) of the binder 2 are equal to a quarter of those of the binder 1 ($\sigma_{1t} = \sigma_t$, $\epsilon_{1t} = \epsilon_t$) if the binder 1 and the binder 2 have the same tensile stiffness. The total tensile load ($P$) is supported by two of the binder 1 and four of the binder 2, and described as follows.

$$P = \sigma_{1t} A_t \times 2 + \frac{\sigma_{2t} A_t}{8} \times 4 = \frac{5\sigma_{1t} A_t}{2} = \frac{5\sigma_{1t} A_t}{\sqrt{2}\eta} \tag{25}$$

Therefore, the nominal stress ($\sigma$) and the nominal strain ($\epsilon$) are

$$\sigma = \frac{P}{L^2/\sqrt{2}} = \frac{\sqrt{2}}{L^2} \cdot \frac{5\sigma_{1t} A_t}{\sqrt{2}\eta} = \frac{5\sigma_{1t} A_t}{\eta L^2} = \frac{5\sigma_{1t} A_t}{\eta L^2} \cdot \frac{2a_b L^2}{N} \left(\eta - 4 \sqrt{\frac{3a_m}{4\pi N}}\right) = \frac{5}{4} \cdot \frac{8a_b}{\zeta N} \cdot \sigma_t \tag{26}$$

$$\epsilon = \frac{L}{L/\sqrt{2}} = \frac{\sqrt{2} \epsilon_{1t}}{L} = \eta \epsilon_t \cdot \frac{1}{2} \left(\eta - 4 \sqrt{\frac{3a_m}{4\pi N}}\right) = \frac{1}{2} \cdot \zeta \epsilon_t \tag{27}$$
3.3. Tensile direction \(\langle 112\rangle\) and \(\langle 111\rangle\)

The case of the tensile direction \(\langle 112\rangle\) in the bcc model and the case of the tensile direction \(\langle 111\rangle\) in the fcc model are also similarly and simply obtained from Fig. 4. In the bcc model, the total load \(P\) is supported by four binders whose orientation directions are all the same. The nominal stress \((\sigma)\) and the nominal strain \((\epsilon)\) are described as follows.

\[
\sigma = \frac{2}{3} \cdot \frac{8\alpha_b}{\xi N} \cdot \sigma_t \quad \text{and} \quad \epsilon = \frac{3}{8} \cdot \xi \epsilon_t \tag{28}
\]

In case of the tensile direction \(\langle 111\rangle\) in the fcc model, the total load \(P\) is supported by two of the binder 1 and four of the binder 2. The orientation directions of the binder 1 and the binder 2 are different, but the elongations of them are equal to each other because the angles between the tensile direction and the orientation directions of the binders are equal to each other \((\cos^{-1}\sqrt{\frac{2}{3}})\). The nominal stress \((\sigma)\) and the nominal strain \((\epsilon)\) are described as follows.

\[
\sigma = 2 \cdot \frac{8\alpha_b}{\xi N} \cdot \sigma_t \quad \text{and} \quad \epsilon = \frac{3}{4} \cdot \xi \epsilon_t \tag{29}
\]

Equations (16), (17), (20), (23), (26)–(29) are summarized as follows.

\[
\sigma = \beta_{\sigma} \cdot \frac{8\alpha_b}{\xi N} \cdot \sigma_t \quad \text{and} \quad \epsilon = \beta_{\epsilon} \cdot \xi \epsilon_t \tag{30}
\]

where \(\beta_{\sigma}\) and \(\beta_{\epsilon}\) are constant coefficients determined by the tensile direction of the models. The values of the constant coefficients are summarized in Table 1.

|       | \(\langle 100\rangle\) | \(\langle 110\rangle\) | \(\langle 112\rangle\) | \(\langle 100\rangle\) | \(\langle 110\rangle\) | \(\langle 111\rangle\) |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \(\beta_{\sigma}\) | 1               | 1               | 2/3             | 1               | 5/4             | 2               |
| \(\beta_{\epsilon}\) | 1               | 1/2             | 5/8             | 1               | 1/2             | 3/4             |

Fig. 4  Microscopic models in the tensile direction \(\langle 112\rangle\) and \(\langle 111\rangle\).
4. Static tensile test
4.1. Specimen and experimental apparatus

Figures 5 and 6 show the schematics of the specimens for the static tensile tests, and Table 2 shows constituent materials of the specimens. The specimens are prepared with graphite powder and thermoplastic fluropolymer binder (polyvinylidene fluoride, PVDF) by reference to negative electrodes for LIBs. The mean diameter of the graphite powder is \( d = 5 \mu m \). The compounding weight ratio Graphite : PDVF is from 94 : 6 to 50 : 50, and 0 : 100. The specimens

| Table 2 | Constituent materials of AM film |
|---------|---------------------------------|
| Material | Specification                   |
| Active material (AM) | Graphite | 5 \( \mu m \) diameter solid particle |
| Adhesive agent (binder) | Polyvinylidene fluoride (PVDF) | Solef5130 made by Solvay |
| Solvent | N-methylpyrrolidone (NMP) | Organic solvent |

Fig. 5 Schematic of 100 wt% specimens for static tensile tests (unit: mm). The thickness of the specimens is \( t_s = 0.042–0.057 \) mm before the tensile tests. The 100 wt% PVDF specimens are prepared as straight plates and dumbbell shaped plates. The 100 wt% PVDF specimens are used to obtain the Young’s modulus \( E_b \) and the tensile strength \( \sigma_{t,b} \) of the PVDF.

Fig. 6 Schematic of 6–50 wt% PVDF specimens for static tensile tests (unit: mm). The shape of the 6 wt% PVDF specimens are straight plates, and the other specimens are dumbbell shaped plates. The thickness of the specimens is \( t_s = 0.068–0.57 \) mm before the tensile tests.

Fig. 7 Tensile testing machine for static tensile tests. The specimen is attached with sponges. The tensile load is applied on the specimen until the specimen fractures. The test speed is 0.5 mm/min.
whose compounding weight ratio Graphite : PDVF is 100 − X : X are hereinafter described as X wt% PVDF specimen. The compounding ratio of the 6 wt% PVDF specimens is standard ratio of graphite negative electrodes (Haselrieder et al., 2015; Giménez et al., 2018). The 25 wt% PVDF specimens refer to silicon negative electrode (Li et al., 2018) and NMC (nickel-manganese-cobalt) positive electrodes (Zhu et al., 2018a). The 100 wt% PVDF specimens are used in order to obtain the Young’s modulus ($E_b$) and the tensile strength ($\sigma_t$) of the PVDF. All of the specimens are manufactured under the atmospheric pressure.

As shown in Fig. 6, the 6 wt% PVDF specimens are straight plates because the 6 wt% PVDF specimens are too fragile to be cut to dumbbell shapes. The 6 wt% PVDF specimens are prepared with the two kinds of the width and the length. The other specimens are dumbbell shaped plates. The 20 wt% and 25 wt% specimens are wider and longer than the others because these specimens are still fragile.

The specimens are attached to the tensile testing machine (Shimadzu Co., Ltd., EZ-LXHS whose capacity is 1 kN) shown in Fig. 7. The both ends of the specimens are clamped with sponges. The tensile load is applied on the specimens until the specimens fracture. The test speed is 0.5 mm/min. The thickness of the 100 wt% PVDF specimens is $t_s = 0.042–0.057$ mm, and the thickness of the 6–20 wt% PVDF specimens is $t_s = 0.068–0.57$ mm before the tensile tests.

4.2. Calculation procedure of tensile strength

The tensile strength of the AM film (6–50 wt% specimens) is calculated by dividing the ultimate load ($P_B$) by the cross-section area of the specimens ($b_s \cdot t_s$). Thus, the experimental values of the tensile strength are calculated as follows.

$$\sigma_B = \frac{P_B}{b_s \cdot t_s} \quad (32)$$

On the other hand, using Eq. (30) and the tensile strength of the binder ($\sigma_{t,B}$), the theoretical values by the proposed models are calculated as follows.

$$\sigma_B = \beta \cdot \frac{8 a_b}{N} \cdot \sigma_{t,B} \quad (33)$$

The tensile strength of the binder ($\sigma_{t,B}$) is obtained by the tensile tests of the 100 wt% PVDF specimens. The comparison of the experimental values (Eq. (32)) and the theoretical values (Eq. (33)) is shown in the next section.

4.3. Calculation procedure of volume ratios

It is difficult to directly measure the volume ratios of the graphite and the PVDF ($\alpha_a$, $\alpha_b$). In the static tensile tests, the volume ratios of the specimens are calculated from the weight ratio of the PVDF ($w_b$) by the following equations.

$$\alpha_a = \frac{m_s (1 - w_b)}{\rho_a V_s} \quad (34)$$

$$\alpha_b = \frac{m_s w_b}{\rho_b V_s} \quad (35)$$

where $\rho_a$ and $\rho_b$ are the mass densities of the graphite and the PVDF. The mass densities ($\rho_a$, $\rho_b$) have been measured before manufacturing the specimens. These mass densities are $\rho_a = 2.26 \times 10^3$ kg/m$^3$ and $\rho_b = 1.78 \times 10^3$ kg/m$^3$.

$m_s$ and $V_s$ are the total mass and the total volume of each specimen. The mass ($m_s$) is measured before the tensile tests. In Eq. (34) and Eq. (35), $m_s (1 - w_b) / \rho_a$ and $m_s w_b / \rho_b$ respectively mean the volumes of the graphite and the PVDF in the specimens. The volume ($V_s$) is computed by multiplying the area of the specimens shown in Fig. 6 by the thickness ($t_s$). For example, the volume of the 6 wt% PVDF specimens is computed as follows.

$$V_s = b_s \cdot L_s \cdot t_s \quad (36)$$

4.4. Results and discussion

Figure 8 shows the stress-strain (S-S) curves of the 100 wt% PVDF specimens, and the fracture process of the specimen No. 5 during the tensile test is shown in Fig. 9 as an example. Using the load $P$ and the elongation $\lambda$ measured in the tensile tests, the nominal stress ($\sigma$) and the nominal strain ($\epsilon$) have been calculated by the following equations.

$$\sigma = \frac{P}{b_s \cdot t_s} \quad (37)$$

$$\epsilon = \frac{\lambda}{L_s} \quad (38)$$
From the results of the specimens No. 1–3, the Young’s modulus of the PVDF \( (E_b) \) is 1 GPa as shown as the black dashed line (linear approximation). The peak stress appears at the crack initiation of the specimens, and seems to be the tensile strength \( (\sigma_t, B) = 25–35 \text{ MPa} \).

As shown in Fig. 8, all of the specimens have a linear relationship at the beginning of the S-S curves. The gradient at the beginning of the S-S curves of the specimens No. 1–3 leads that the Young’s modulus of the PVDF \( (E_b) \) is 1 GPa. This value agrees with the reference value 1–1.5 GPa (Solvay, 2019). At the nominal strain \( \varepsilon = 0.07–0.08 \), the stress peak appears in most of the specimens. As shown in Fig. 9, one crack initiated in the specimens at the stress peak. After the crack initiation, the stress \( (\sigma) \) decreases with the increase of the strain \( (\varepsilon) \). In the specimens, multiple cracks occurred, and the cracks grew to voids. The voids merged into a single void, and the specimens finally fractured. At the first crack initiation i.e. the stress peak, there is no change in the width \( (b_s) \) and the thickness \( (t_s) \) in all of the specimens. Therefore, the tensile strength of the PVDF \( (\sigma_{t,B}) \) seems to be 25–35 MPa.

Figure 10 shows the volume ratios of the graphite and the PVDF \( (\alpha_a, \alpha_b) \) subjected to the weight ratio of the PVDF \( (w_b) \) in the 6–50 wt% PVDF specimens. The volume ratio of the graphite is constant \( (\alpha_a = 35.3 \%) \), and the volume ratio of the PVDF is proportional to the weight ratio of the PVDF \( (\alpha_b = 0.77 \text{ wt%}) \). The residual is the volume ratio of pores in the specimens. Even if the volume ratio of the PVDF \( (\alpha_b) \) increases, the volume ratio of the graphite \( (\alpha_a) \) is constant because the solved PVDF binder seems to locate at pores among graphite particles in the manufacturing process of specimens. The specimens were manufactured under the atmospheric pressure in this study. Some other manufacturing methods apply pressure during the drying process in order to control the porosity. Thus, the volume ratio of the graphite \( (\alpha_a) \) will be varied by the pressure. The values shown in Fig. 10 are used for the calculation of the theoretical values of the tensile strength \( (\sigma_{t,B}) \). The parameters for the calculation of the theoretical values are summarized in Table 3.

Figure 11 shows examples of the 6 wt% specimens and the 30 wt% specimens after the tensile tests. The macroscopic fracture of the specimens is brittle fracture. The decreases of the width \( (b_s) \) and the thickness \( (t_s) \) of the specimens are
Volume ratios of graphite and PVDF subjected to weight ratio of PVDF in the 6–50 wt% PVDF specimens.

The volume ratio of the graphite is constant, and the volume ratio of the PVDF is proportional to the weight ratio of the PVDF.

![Graph](image1)

Fig. 10 Volume ratios of graphite and PVDF subjected to weight ratio of PVDF in the 6–50 wt% PVDF specimens. The volume ratio of the graphite is constant, and the volume ratio of the PVDF is proportional to the weight ratio of the PVDF.

![Graph](image2)

Table 3 Summary of parameters for calculation of theoretical value of tensile strength (%)

|          | bcc model | fcc model |
|----------|-----------|-----------|
| Volume ratio of PVDF, α_b | Calculated by α_b = 0.77 w_b | 0.353 |
| Volume ratio of graphite, α_a | 0.353 |
| Number of particles in unit cube, n | 2 | 4 |
| Constant coefficient, η | √3 | √2 |
| Constant coefficient (Eq. (18)), ζ | 0.589 | 0.438 |
| Coordination number, N | 8 | 12 |
| Tensile strength of PVDF, σ_{t,B} | 30 MPa (average of 25–35 MPa) |

![Image](image3)

(a) 6 wt% PVDF specimen

![Image](image4)

(b) 30 wt% PVDF specimen

Fig. 11 Pictures of examples of the 6 wt% specimens and the 30 wt% specimens after the tensile tests (left: macroscopic pictures, right: SEM images). The macroscopic fracture of the specimens is brittle fracture. The decreases of the width and the thickness are small. The SEM images show the fracture surfaces of the specimens. The white arrows indicate the fracture of the PVDF binder which bonded the graphite particles.

small enough to be ignored between before and after the tensile tests. The SEM images show the fracture surfaces of the specimens. The white arrows indicate the fracture of the PVDF binder which bonded the graphite particles. Many marks of fractured binder were observed by the SEM. Fracture of graphite particles or debonding between binder and graphite particles were not observed. In this respect, the binder clearly support the structure of the AM film.

Figure 12 shows the experimental values and the theoretical values of the tensile strength of the 6–50 wt% PVDF specimens. The experimental values are calculated by Eq. (32). The cross-section area \( b_s \cdot t_s \) before the tensile tests is used because the decreases of the width \( b_s \) and the thickness \( t_s \) of the specimens can be ignored as described in the

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Fig. 12 Tensile strength of 6–50 wt% specimens. The tensile strength increases with the increase of the volume ratio of the PVDF. Most of the experimental values locate among the theoretical lines of the two models in the tensile direction $<100>$ and $<110>$. Some of the specimens with low strength are close to the bcc model in the tensile direction $<112>$. Therefore, the PVDF supports the structure of the AM film, and both the bcc model and the fcc model can simulate the structure of the AM film.

previous paragraph. The theoretical values are calculated by Eq. (33) and the parameters shown in Table 3. As shown in Fig. 12, the experimental values of the tensile strength ($σ_B$) increase with the increase of the volume ratio of the PVDF ($α_b$). All of the theoretical values linearly increase with the volume ratio of the PVDF.

Most of the experimental values locate between the theoretical lines of the fcc model in the tensile direction $<100>$ and $<110>$. The theoretical lines of the bcc model in the tensile direction $<100>$ and $<110>$ are between the fcc model in the tensile direction $<100>$ and $<110>$, and some of the experimental values agree with the theoretical lines of the bcc model in the tensile direction $<100>$ and $<110>$. These results mean that most of the specimens can be simulated by both the bcc model and the fcc model in the tensile direction $<100>$ and $<110>$. Some of the specimens with low strength are close to the theoretical lines of the bcc model in the tensile direction $<112>$. In this respect, the bcc model can estimate the lowest strength of the specimens. None of the specimens achieve the theoretical strength of the fcc model in the tensile direction $<111>$ which has the highest strength. Therefore, both the bcc model and the fcc model can simply simulate the structure of the AM film, and the difference of the tensile direction of the models can estimate the variability of the tensile strength of the AM film.

5. Bending fatigue test

5.1. Specimen and experimental apparatus

There is difficulty to attach AM film on tensile testing machines in that the AM film is too fragile to be clamped by chucks. For this reason, this study uses a specimen in which AM film coated on copper foil, and applies bending fatigue tests on the specimen. Figure 13 shows the schematic of the specimen for the bending fatigue tests. The constituent materials of the AM film are the same as shown in Table 2. The compounding weight ratio Graphite : PVDF is 94 : 6. The AM film is manufactured under the atmospheric pressure as with the static tensile tests. The thickness of the AM film is $t_a = 0.18–0.45$ mm before the bending tests. The thickness of the copper foil is $t_c = 0.05$ mm (pure copper, C1220) and 0.1 mm (pure copper, C1100). Strain gauges are attached to the points A to F on the surface of the copper foil in order to measure the strain of the specimen.

Figure 14 shows the bending fatigue testing machine developed in this study. The specimen is attached on the curved plate whose curvature radius is 50 mm. The arm flaps, and the specimen is bended repeatedly while the motor rotates. The rotating speed of the motor is set to 5 Hz. The strain gauges measure the strain of the surface of the copper foil during the tests. The camera is set above the specimen and the behavior of the specimen is observed during the tests. Then the number of cycles ($N_f$) of the bending until the crack initiation on the AM film is measured.

5.2. Calculation procedure of stress amplitude

The stress amplitude of the binder ($Δσ_r/2$) is calculated by using the maximum strain ($ε_{max}$) and the minimum strain ($ε_{min}$) of the surface of the copper foil, the thicknesses of the AM film and the copper foil ($t_a$, $t_c$) and the curvature radius of the curved plate (50 mm). The calculation uses the maximum strain ($ε_{max}$) and the minimum strain ($ε_{min}$) at the measurement point closest to the macroscopic crack initiated on the AM film. The calculation procedure is as follows.
Figure 13 Schematic of specimen for bending fatigue tests (unit: mm). The specimen is AM film composed of graphite and PVDF coated on copper foil. Strain gauges are attached to the points A–F on the surface of the copper foil in order to measure the strain of the specimen.

Figure 14 Bending fatigue testing machine developed in this study (unit: mm). The arm flaps, and the specimen is bended repeatedly during the motor rotates. The camera set above the specimen observes the behavior of the specimen.

Figure 15 shows the schematic of the specimen in the loading process. The strain is composed of pure tensile strain and bending strain. In addition, the neutral axis of the bending strain is presumed to be at the central axis of the copper foil because the Young’s modulus of copper ($E_c = 118$ GPa) is much larger than that of PVDF ($E_b = 1$ GPa) which mainly supports the construction of the AM film. Based on Fig. 15, the maximum strain ($\epsilon_{a,max}$) of the surface of the AM film is described as follows.

$$\epsilon_{a,max} = \epsilon_{max} - \frac{t_c/2}{\rho_{max}} + \frac{t_c/2 + t_a}{\rho_{max}} = \epsilon_{max} + \frac{t_a}{\rho_{max}} \tag{39}$$

The maximum strain of the copper foil ($\epsilon_{max}$) and the maximum curvature radius of the specimen ($\rho_{max}$) occur when the specimen enough contacts the curved plate. Therefore, the maximum curvature radius ($\rho_{max}$) is equal to the curvature radius of the curved plate ($\rho_{max} = 50$ mm).

Similarly, the minimum strain ($\epsilon_{a,min}$) of the surface of the AM film is described as follows.

$$\epsilon_{a,min} = \epsilon_{min} - \frac{t_c/2}{\rho_{min}} + \frac{t_c/2 + t_a}{\rho_{min}} = \epsilon_{min} + \frac{t_a}{\rho_{min}} \tag{40}$$

The minimum strain of the copper foil ($\epsilon_{min}$) and the minimum curvature radius of the specimen ($\rho_{min}$) occur when the specimen leaves the curved plate. Taking account of that the pure tensile strain is zero, the minimum curvature radius ($\rho_{min}$) is described as follows.

$$\rho_{min} = \frac{t_c}{2\epsilon_{min}} \tag{41}$$
Then, Eq. (40) is described as follows.

$$\epsilon_{a,max} = \left(1 + \frac{2t_b}{t_c}\right)\epsilon_{\text{min}}$$  \hspace{1cm} (42)

Hence, the stress amplitude ($\Delta \sigma_t/2$) of the binder are calculated by using Eq. (31) and the Young’s modulus of the binder ($E_b$) as follows.

$$\frac{\Delta \sigma_t}{2} = \frac{E_b(\epsilon_{\text{max}} - \epsilon_{\text{min}})}{2\beta_x \zeta} = \frac{E_b}{2}\left(\frac{\epsilon_{\text{max}} + \frac{t_a}{t_{\text{max}}} - \left(1 + \frac{2t_b}{t_c}\right)\epsilon_{\text{min}}}{\beta_x \zeta}\right)$$  \hspace{1cm} (43)

5.3. Results and discussion

Figure 16 shows an example of the crack initiation of the AM film (the number of cycles $N_f = 4.8 \times 10^4$). All of the cracks initiated on the surface of the AM film, and there was no delamination between the AM film and the copper foil. Macroscopically, the cracks initiated and propagated vertically to the longitudinal direction of the specimens in 1 cycle and seemed to be brittle fracture. Turning to the strain measurement, all of the cracks were located close to the measurement points C or D shown in Fig. 13. The maximum strain ($\epsilon_{\text{max}}$) and the minimum strain ($\epsilon_{\text{min}}$) at these measurement points were used to calculate the stress amplitude ($\Delta \sigma_t/2$).

Figure 17 shows the S-N curve of the AM film obtained by the bending fatigue tests. The stress amplitude ($\Delta \sigma_t/2$) is calculated by Eq. (43). From the results of the static tensile tests, the Young’s modulus ($E_b$) is set to 1 GPa, and the constant coefficient ($\zeta$) is set to 0.5 that is average of 0.589 in the bcc model and 0.438 in the fcc model. The constant coefficient ($\beta_x$) is set to 0.75 that is average of Table 1. From the results of the bending fatigue tests, the stress ($\Delta \sigma_t$) at the 1 cycle is 23–31 MPa and this value is close to the tensile strength of the PVDF (25–35 MPa) as shown in Fig. 8. Taking account of the variation of the Young’s modulus in the reference value ($E_b = 1–1.5$ GPa) and the constant coefficient.
the difference of the stress at the 1 cycle and the tensile strength of the PVDF in the tensile tests is within the margin of variation. This means that the PVDF binder supports the AM film, and the proposed models can estimate the stress applied on the binder also in the bending fatigue tests.

Furthermore, the number of cycles ($N_f$) linearly increases in the log scale with the decrease of the stress amplitude ($\Delta \sigma_t/2$). The crack of the AM film is initiated by the $10^6$–$10^7$ cycles at the half of the tensile strength of the PVDF. These tendencies seem to be the fatigue property of the PVDF binder in the AM film, and similar to fatigue properties of usual materials such as aluminum alloy and copper alloy. Therefore, it is believed that the proposed models and the bending tests successfully evaluate the mechanical fatigue of the AM film. The further study including investigations of different composition ratio of binder and different constituent materials such as positive electrodes is an issue in the near future.

6. Conclusions

This study has proposed a simple evaluation method of mechanical strength and fatigue property of electrodes for LIBs by using the mechanical models of the electrodes. The proposed models approximate the alignment of particles of AM as the body-centered cubic (bcc) and the face-centered cubic (fcc) which are the well-known crystal lattices. Binder is approximated to be elastic bars that joint the particles. In order to verify the proposed method, static tensile tests and bending fatigue tests of negative electrodes for LIBs have been conducted. The followings have been summarized from the test results.

(1) In the static tensile tests, the macroscopic fracture of the specimens is brittle fracture. The microscopic observation reveals that the binder clearly supports the structure of the AM film. The tensile strength of the specimens estimated by the proposed models agree with the experimental values. Both the bcc model and the fcc model can simply simulate the structure of the AM film, and the difference of the tensile direction of the models can estimate the variability of the tensile strength of the AM film.

(2) In the bending fatigue tests, all of the cracks initiated on the surface of the AM film. The macroscopic cracks initiate and propagate in 1 cycle and seem to be brittle fracture. The estimation value of the stress that initiates the cracks on the specimens by 1 cycle agrees with the tensile strength. The number of cycles linearly increases in the log scale with the decrease of the stress amplitude, and the stress amplitude at the $10^6$–$10^7$ cycles agrees with the half of the tensile strength. These tendencies seem to be the fatigue property of the binder in the AM film, and similar to fatigue properties of usual materials. Therefore, it is believed that the proposed models and the bending fatigue tests successfully evaluate the mechanical fatigue of the AM film.

The further study including investigations of different constituent materials such as positive electrodes and development of the mechanical models is an issue in the near future. This work was supported by JSPS KAKENHI Grant Numbers JP16K05989 and JP19K04078. The microscopic observation of the specimens was supported by Interdisciplinary Research Center for Nano Science and Technology in Tokyo City University. The authors would like to thank Tatsuya Tsuruta, Hitoshi Aoki, Kazunari Hashimoto and Naoya Yamaoka, students at Tokyo City University. The basic of this work is supported by their contribution.

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