Original Papers

Design of Ceramic Materials with Strong Adhesion to Resin with a Combination of Orthogonal Array and Response-Surface Method

by

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Appropriate ceramic materials with strong adhesion to a resin, which is used for thin-film devices, were selected by using a combination of an orthogonal array and a response-surface method. In this technique, at the first step, important factors that significantly influence the adhesion strength were selected from various factors that characterize ceramic materials by using an orthogonal array with molecular simulations. As a result, the short-side and long-side lattice constants \(a\) and \(b\) were selected from four ceramic-material factors \(a\), \(b\), the surface energy density, \(S\), and the cohesive energy, \(C\). At the second step, the adhesion strength was described as a function of the selected important factors by using a response-surface method. From this function, the ideally most appropriate values for \(a\) and \(b\) that made the adhesion strength maximum were obtained. The ideal values for \(a\) and \(b\) were obtained as 0.245 nm and 0.424 nm, respectively. At the third step, the most appropriate ceramic material whose lattice constants were the closest to the ideal values \(a = 0.245\) nm and \(b = 0.424\) nm was selected by use of the simulation results of lattice constants. As a result, a \(\text{SiO}_2/\text{Al}_2\text{O}_3/\text{TiO}_2\) multilayer whose lattice constants were \(a = 0.243\) nm and \(b = 0.421\) nm (the closest to the ideal values) was selected as the most appropriate ceramic material with the strongest adhesion to the resin.

Key words:

Adhesion strength, Ceramic materials, Resin, Response-surface method, Molecular simulation, Orthogonal array

1 Introduction

Recently, hybrid organic-inorganic materials (e.g., composite materials and compounded resins) are used to improve the performance and reliability of thin-film devices such as biomechanics devices, semiconductor packages, magnetic recording media, and optical discs. Because the devices are expected to be used at places of high temperatures and high humidity in the upcoming IoT (Internet of Things) era, the interface adhesion between organic and inorganic materials needs to be strong to realize the sufficient device reliability. When the interface between the organic and inorganic materials is not stable, adhesive fractures occur at the interface at high temperatures or in high humidity. Selecting appropriate materials is one of the keys to preventing the adhesive fractures.

In the field of electronic packaging, for example, it is well known that the adhesion strength of electroplated nickel thin-film interconnections to chromium underlayers is very strong. Generally, however, it is difficult to select an appropriate material that has strong adhesion to another material by using a conventional trial-and-error approach. So, recently many researchers have paid attention to materials-informatics techniques, which can select an appropriate material by using information science (for example, response-surface methods\(^1\)\(^2\)) with computer simulation and experimental data. However, the response-surface methods sometimes need a long time to obtain results because they usually use a lot of data and parameters (materials factors). For example, in Refs. 1) and 2), a response-surface method was applied to optimize materials properties by use of a lot of data, which needed much time to analyze. So, by combining an orthogonal array\(^3\) with the surface-response method\(^1\)\(^2\), the author has developed a technology for efficiently selecting an inorganic material with strong adhesion to an organic material (resin). This paper describes the application of this technology to the selection of a ceramic material that has strong adhesion to a resin, while in another paper, the metal that has the strong adhesion to a resin is selected by using this efficient technology. In the field of thin-film electronics devices, ceramic materials are used as filler particles or fibers in resins to improve resin properties such as the mechanical strength, insulating properties, and thermal conductivity, etc. When the adhesive fracture occurs at the interface between ceramic materials and the resin in electronics devices, mechanical or electrical failures are caused. So it is important to select appropriate ceramic materials with strong adhesion to resins.

In the technology that the author has developed, at the first step, the important factors that significantly influence the adhesion strength are selected from ceramic-material factors such as lattice constants and surface energy density that characterize ceramic materials by using an orthogonal array\(^3\) and molecular simulations\(^4\). At the second step, by using a response-surface method\(^1\)\(^2\), the adhesion strength is described as a function of the selected important factors, and the most appropriate values of the factors that make the adhesion strength maximum are obtained. At the third (final) step, appropriate ceramic materials whose factors are close to the most appropriate values are selected.

In this study, the author used an alicyclic-ring-based
resin as an example because the resin is often used in electronics devices and lithium-ion batteries alongside an aromatic-ring-based resin, which was described in another paper. Then, the best ceramic material with strong adhesion to this alicyclic-ring-based resin was selected.

2 Material-Selection Technique

2.1 Material-Selection Technique Made of Three Steps

The basic idea of material selection in the author’s technique shown in Fig. 1 is to select a material whose factors \( A, B, C, \ldots \) are close to the most appropriate values \( A_0, B_0, C_0, \ldots \) that optimize an objective material property, \( F \), by describing the property as a function of material factors (i.e., \( F(A, B, C, \ldots) \)). In this study, the objective material property, \( F \) that the author wants to optimize is the adhesion to a resin, and the factors \( A, B, C, \ldots \) that the author focus on are the lattice constants (geometrical factors), the surface energy density, and the cohesive energy of a ceramic material (energetic factors). Although other factors such as surface steps, oxygen defects, and interface roughness generally influence the adhesion, these factors are neglected in this paper because this paper’s application devices are thin-film devices in which few defects and little interface roughness exist, as shown in an example TEM image (Fig. 2).

To describe the material property \( F \) as a function of material factors \( A, B, C, \ldots \), a number of data of experiments or simulations to interpolate are needed. If too many factors are used, many data are needed for interpolation and it takes too much time to obtain the function \( F(A, B, C, \ldots) \). So the author uses orthogonal-array analysis\(^3\) to select only the important factors that strongly influence the adhesion (objective material property).

The overview of the material-selection technique is shown in Fig. 1. This figure shows that the method consists of three steps. At the first step, by using the orthogonal-array sensitivity analysis\(^3\) with molecular simulations\(^6\), the important material factors that significantly influence an objective material property are selected from material factors (the lattice constants, the surface energy density, and the cohesive energy) that characterize ceramic materials. In the sensitivity analysis, the results of molecular simulations of the objective material property that is paid attention to are used as input data. At the second step, the objective material property, which is the adhesion in this paper, is described as a function of the selected important factors by using a response-surface method\(^3\) with the molecular-simulation results that are also used at the first step. By using this function, the most appropriate values of the factors that optimize the objective material property, which is adhesion strength in this paper, are determined. At the third (final) step, the materials (ceramic materials, in this paper) whose factors are close to the most appropriate values are selected.

![Fig. 2 TEM image of an interface between thin films](image_url)

![Fig. 3 Schematic structure of an alicyclic-ring-based resin.](image_url)

2.2 Simulations Used at the First and Second Steps

As shown in Fig. 1, molecular simulations are used in the first and second steps. So the method of the simulations is explained in this section. In this paper, an example resin (Fig. 3) that contains alicyclic rings is used because this resin is often used in electronics devices for the reason that this has strong mechanical strength and heat resistance. The simulation model of this example resin is shown in Figs. 3 and 4. Figure 3 shows a two dimensional schematic structure
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of the resin, which is not a stabilized structure. Figure 4 shows the stabilized structure of the resin. The simulation model that the author used in this study is an interface between the resin (Fig. 4) and a ceramic multilayer, as shown in Fig. 5. By using the ceramic multilayer made up of atomic-scale thin films shown in Fig. 5, instead of using a single ceramic material, it was possible to control the lattice constants of the top layer shown in Fig. 6 almost freely. This simulation model was made by using "Materials Studio®" software from Dassault Systemes BIOVIA.

Sixteen resin molecules (four layers of four molecules), one of which is shown in Fig 4, were used as the resin film model. There are 2,368 atoms in the resin molecules.

Next, the size of the ceramic-multilayer model shown in Fig. 5 is explained in this paragraph. From the past molecular-dynamics simulations\(^1\) it was found that the adhesion strength between the resin and inorganic materials was almost independent of the thickness when the thickness was larger than 2 nm. So, in this study, the author set the thickness of the ceramic layer at 2.4 nm. The values of thickness of Ceramics 1, 2, and 3 in Fig. 5 are set at 0.6 nm, 0.9 nm, and 0.9 nm, respectively. These values were determined to make it easy to control the lattice constants of the top layer of the ceramic multilayer. The sizes of the ceramics in the \(x\) and \(y\) directions are set at 3.2 nm and 5.2 nm, which are large enough for attachment of sixteen resin molecules (four layers of four molecules).

In this study, the adhesion strength was evaluated by calculating the adhesive fracture energy\(^3\) that was defined as the difference between the area density of potential energy of the resin/ceramics-attached state (Fig. 5) and that of the resin/ceramics-detached state (Fig. 7). The fracture energy, as well as the surface energy density defined as half of the difference between the area density of potential energy of Fig. 8 (a) and that of Fig. 8 (b), was calculated by using molecular dynamics with a universal force field\(^5\).\(^6\). The temperature was kept at 20°C by using a velocity-scaling method\(^7\).

As described in Refs. 5) and 6), the universal force field used in molecular-dynamics simulations is determined based on the element, its hybridization, and its connectivity. As explained in these references, the potential energy of this force field is expressed as a sum of valence or bonded
interactions and nonbonded interactions in the following equation.

\[ P = P_k + P_o + P_e + P_{\text{vdw}} + P_{\text{el}} \]  (1)

Here, bond stretching \( P_k \), bond angle bending \( P_o \), dihedral angle torsion \( P_e \), and inversion terms \( P_{\text{vdw}} \) are valence interactions. On the other hand, van der Waals \( P_{\text{vdw}} \) terms and electrostatic \( P_{\text{el}} \) terms are nonbonded interactions. The universal force field expressed by Eq. (1) was employed in this study because the results obtained with this force field were confirmed to agree well with the results obtained with first-principle quantum simulations (density-functional-theory simulations) by using the software, Dmol3, from Dassault Systemes BIOVIA. The results of the quantum simulations will be discussed in the next paper.

The electrostatic \( P_{\text{el}} \) terms in Eq. (1) are long-range Coulomb interactions induced by the partial charges. So, the atoms were kept free to move, and no periodic boundary conditions are set so that long range electrostatic forces from the replica molecular models induced by the periodic potential energy of the surface-connected state (Fig. 8 (a)) and that of the surface-separated state (Fig. 8 (b)). The surface energy density, \( S \) was calculated by using molecular dynamics with a universal force field\(^5\), \(^6\).

The cohesive energy, \( C \), is defined as the difference between the potential energy of the free atoms and that of the atoms of a solid.

### 2.4 Orthogonal Array Used at the First Step

At the first step of the material-selection technique (Fig. 1), the important material factors (ceramic factors) that significantly influence the objective material property (adhesion strength) are selected by clarifying the dependence of the adhesion strength on the four ceramic factors. When we investigate the dependence, we usually change the value of one factor with the other three factors fixed. However, it is time-consuming to clarify the dependence by using this method. Accordingly, the author used the orthogonal-array design methodology\(^3\), which is known to be effective for efficiently designing macro-scale structures such as propeller fans and diaphragms. In this methodology, when there are four material factors, only nine simulations or experiments shown in an L9 orthogonal array (see Table 1) are needed to clarify the important factors on which the adhesion strength strongly depends\(^3\). Here the author set Level 1 (small values), Level 2 (middle values), and Level 3 (large values) of the four factors at the regions shown in Table 2. The author set these levels so that each level has about one third of the important factors selected by use of the orthogonal-array analysis explained in Section 2.4. In this study, the author employed a response-surface method\(^1\), \(^2\), so that all the data points used for interpolation are one-dimensional data interpolation by use of the Kriging method. Accordingly, the author used the orthogonal-array design methodology\(^3\), which is known to be effective for efficiently designing macro-scale structures such as propeller fans and diaphragms. In this methodology, when there are four material factors, only nine simulations or experiments shown in an L9 orthogonal array (see Table 1) are needed to clarify the important factors on which the adhesion strength strongly depends\(^3\). Here the author set Level 1 (small values), Level 2 (middle values), and Level 3 (large values) of the four factors at the regions shown in Table 2. The author set these levels so that each level has about one third of the important factors selected by use of the orthogonal-array analysis explained in Section 2.4.
multilayers in Table 1 (i.e., Table 3), the sensitivity of Level \( j \) of the \( i \)-th factor, \( W_j \) is given by
\[
W_j = 10 \log \left( \frac{P_j - Q_j}{n} \right),
\]
where
\[
P_j = (D_{i1}^2 + D_{i2}^2 + \cdots + D_{in}^2)/n
\]
and
\[
Q_j = (D_{1i}^2 + D_{2i}^2 + \cdots + D_{ni}^2 - P_j n)/n - 1.
\]
Here, \( D_{ij} \) is the \( i \)-th force field-energy datum that belongs to Level \( j \) of the \( i \)-th factor, and \( n \) is the number of the data that belong to Level \( j \) of the \( i \)-th factor.

**Table 1** L9 orthogonal array in the case of four factors.

| Condition | 1st factor: Short-side lattice constant \( a \) (\( \text{nm} \)) | 2nd factor: Long-side lattice constant \( b \) (\( \text{nm} \)) | 3rd factor: Surface energy density \( S \) (\( \text{J} / \text{m}^2 \)) | 4th factor: Cohesive energy \( C \) (\( \text{J} / \text{mol} \)) |
|-----------|---------------------|---------------------|---------------------|---------------------|
| Level 1   | Level 1             | Level 1             | Level 1             | Level 1             |
| Level 2   | Level 1             | Level 2             | Level 2             | Level 2             |
| Level 3   | Level 1             | Level 3             | Level 3             | Level 3             |
| Level 4   | Level 2             | Level 1             | Level 2             | Level 1             |
| Level 5   | Level 2             | Level 2             | Level 3             | Level 1             |
| Level 6   | Level 2             | Level 3             | Level 1             | Level 2             |
| Level 7   | Level 3             | Level 1             | Level 2             | Level 2             |
| Level 8   | Level 3             | Level 2             | Level 1             | Level 3             |
| Level 9   | Level 3             | Level 3             | Level 2             | Level 1             |

**Table 2** Factors and levels for ceramic multilayers.

| 1st parameter: Short-side lattice constant \( a \) (\( \text{nm} \)) | 2nd parameter: Long-side lattice constant \( b \) (\( \text{nm} \)) | 3rd parameter: Surface energy density \( S \) (\( \text{J} / \text{m}^2 \)) | 4th parameter: Cohesive energy \( C \) (\( \text{J} / \text{mol} \)) |
|---------------------|---------------------|---------------------|---------------------|
| Level 1 (small)     | \( \beta \) < 0.250 | \( \gamma \) < 0.430 | \( \delta \) < 3.0   |
| Level 2 (middle)    | \( \beta \) > 0.250 | \( \gamma \) > 0.430 | \( \delta \) > 3.0   |
| Level 3 (large)     | \( \beta \) > 0.250 | \( \gamma \) > 0.430 | \( \delta \) > 3.0   |

**Table 3** Nine ceramic multilayers corresponding to Table 1 (L9), where Levels 1, 2, and 3 are shown in Table 2.

According to the sensitivity-analysis method\(^3\) based on orthogonal arrays, the factors whose increase from Level 1 to Level 3 significantly influence the sensitivity \( W_j \) are important in optimizing the adhesion strength, and they are selected as important factors at the first step of the material-selection technique (Fig. 1). At the second step in Fig. 1, the adhesion strength is described as a function of the selected factors by interpolating the nine fracture-energy data corresponding to the L9 orthogonal array in Tables 1 and 3 by use of a response-surface method.

### 2.5 Response-Surface Method

At the second step of the material-selection technique (Fig. 1), a response-surface method is used to describe the objective material property (adhesion strength) as a function of the important factors selected by use of the orthogonal-array analysis explained in Section 2.4. In this study, the author employed a response-surface method\(^1, 2\), so called Kriging method, which is often used in geostatistics and mechanical structure design. The schematic view of one-dimensional data interpolation by use of the Kriging method is shown in Fig. 9. In this method, the function curve is made so that all the data points used for interpolation are on the curve. The basic idea of the Kriging method is to predict the value of a function at a given point by computing a weighted average of the known values of the function in the neighborhood of the point. The theory derives a best linear unbiased estimator, based on assumptions on covariances, and is mathematically related to regression analysis.
values of the important factors (the short-side and long-side lattice constants, nine data in Table 4 with the Kriging method. The obtained constants shown in Fig. 4, it is found that the lattice matching between the ceramic multilayer and the resin is the most important in increasing the adhesion strength between them. Because these values are the same values of resin’s lattice constant, the interface between the ceramic multilayer and the resin is the most important in increasing the adhesion strength between them. At the second step of Fig. 1, the author described the adhesive fracture energy as a function by interpolating the nine data in Table 4 with the Kriging method. The obtained function is shown in Fig. 11. The ideally most appropriate values of the important factors (the short-side and long-side lattice constants, a and b) are obtained as a = 0.245 nm and b = 0.424 nm by using the gradient of the obtained function. Because these values are the same values of resin’s lattice constants shown in Fig. 4, it is found that the lattice matching between the ceramic multilayer and the resin is the most important in increasing the adhesion strength between them. At the third (final) stage of Fig. 1, the appropriate materials (ceramic multilayers) whose factors are close to the most appropriate values (a = 0.245 nm and b = 0.424 nm, which are resin’s lattice constants) are selected. The author selected SiO2/Al2O3/TiO2 multilayer because the molecular simulation showed that the lattice constants of this ceramic multilayer are [a = 0.243 nm and b = 0.421 nm] and that these values are close to the resin’s lattice constants. By calculating the adhesive fracture energy defined in Section 2.2, the value for the SiO2/Al2O3/TiO2 multilayer was obtained as 0.381 J/m², which is higher than all values shown in Table 4. To clarify the reason that SiO2/Al2O3/TiO2 has strong adhesion to the resin, the author visualized the atomic configuration of the interface between the resin and SiO2/Al2O3/TiO2, as shown in Fig. 12. Because Fig. 12 shows that oxygen atoms of the top SiO2 layer can be seen through near the centers of the alicyclic rings and that the configuration of the oxygen atoms are in harmony with alicyclic rings and alkyl chains, the accumulation of atomic interactions caused by this harmony is considered to lead to the strong adhesion.

The atomic configurations of the interface between the resin and SiO2/Al2O3/TiO2 whose adhesive fracture energy (0.252 J/m²) is smaller than that of the resin/SiO2/Al2O3/TiO2 interface are shown in Fig. 13. This figure shows that oxygen atoms can not be seen through near the alicyclic-ring centers. Furthermore, the lattice matching configuration of the oxygen atoms, alicyclic rings, and alkyl chains, which is seen in Fig. 12, is not seen in Fig. 13. Accordingly, carbon atoms in the resin are not positioned at energetically stable sites in Fig. 13 in spite of high surface energy and high cohesive energy. So the accumulation of atomic interaction at the interface between the resin and SiO2/Al2O3/TiO2 is smaller than that at the interface shown in Fig. 12. Thus, the accumulation of atomic interactions caused by the lattice matching seen at the interface between the resin and SiO2/Al2O3/TiO2 (Fig. 12) is confirmed to lead to the strong adhesion.
By using the 0-2θ scheme of the X-ray diffraction (XRD) analysis, the author selected the specimens that have significant diffraction peaks, which show single-orientation films. An example result of the XRD for SiO2/Al2O3/TiO2 is shown in Fig. 14.

![Fig. 14 X-ray diffraction of a SiO2/Al2O3/TiO2 film.](image)

### 4 Comparison between Simulations and Experiments

To confirm the effectiveness of the molecular-dynamics simulation technique described in Chapters 2 and 3, the author conducted a scratch test on the film-laminated structure (scratch tester: CSR-02 made by Rhesca Co., Ltd.) and compared the test results with the simulation results. Recently, more advanced methods for measuring adhesion strength have been proposed in a number of research papers. However, these advanced methods were very difficult to apply to fairly strong interfaces such as the interface between the resin and SiO2/Al2O3/TiO2. Accordingly, a simple scratch-testing method that was applicable to the resin/SiO2/Al2O3/TiO2 interface was employed. In the scratch testing, a diamond indenter with a tip radius of 5 µm makes the dynamic indentations. During the measurement, load is gradually increased and when it reaches a critical load, adhesive fracture occurs at the interface. The adhesion strength is determined by measuring the critical load, L, at which the adhesive fracture occurs.

The author used 0.01-mm-thick naphthalene-based resin films deposited on the ceramics for the scratch test. The molecular structure of the resin films is the same as that in Fig 4. For the ceramics, the author used the five ceramic multilayers (SiO2/Al2O3/TiO2, SiO2/SnO2/GeO2, SiO2/TiO2/BaO2, TiO2/ZrO2/RuO2, and TiO2/HfO2/BaO2), four of which are listed in Table 4. These ceramic multilayers were deposited on silicon substrates by using physical vapor deposition (PVD), and the thickness of each film was set at the same as that used in the simulations by controlling the deposition time so that misfit dislocations were not formed.
the SiO₂/Al₂O₃/TiO₂ multilayer, which was selected by using our material-selection technique (Fig. 1), has the strongest adhesion to the resin. Thus, our technique for selecting appropriate materials with a combination of an orthogonal array and a response-surface method is considered to be effective for selecting a ceramic material that has strong adhesion to the resin.

5 Summary
A technique for efficiently selecting a ceramic material with strong adhesion to a resin with a combination of an orthogonal array and a response-surface method was used to select appropriate ceramics for thin-film devices. In this technique, at the first step, important factors that significantly influence the adhesion strength to the resin were selected from various factors that characterize ceramics by use of an orthogonal array with molecular simulations. At the second step, the adhesion strength was described as a function of the selected important factors by using a response-surface method (Kriging method). From this function, the most appropriate values of the important factors that made the adhesion strength maximum were obtained. At the third step, the most appropriate ceramic materials whose important factors were close to the most appropriate values were selected. By carrying out these procedures on an alicyclic-ring-based resin, which is often used in thin-film devices, the following results were obtained.

(1) By using the sensitivity analysis based on an orthogonal array, the short-side and long-side lattice constants, a and b were selected as the important factors from four ceramic-multilayer factors (the lattice constants, a, b, the surface energy density, S, and the cohesive energy, C).

(2) By describing the adhesive fracture energy as a function of the important factors (the short-side and long-side lattice constants, a and b), the ideally most appropriate values for a and b that made the adhesion strength maximum were obtained as 0.245 nm and 0.424 nm, respectively.

(3) The most appropriate ceramic multilayer whose lattice constants were close to a =0.245 nm and b=0.424 nm (the ideal values) was selected. As a result, a SiO₂/Al₂O₃/TiO₂ multilayer whose lattice constants were a =0.243 nm and b=0.421 nm was selected as the most appropriate ceramic with the strongest adhesion to an example resin (alicyclic-ring-based resin).

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