

Metal Surface Preparation Technology for Adhesive Joining Reliability

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Abstract

In recent years, adhesive bonding has attracted interest as a key joining technology for lightweight metals and dissimilar materials, and its importance is increasing across various industrial fields.

When bonding metallic materials, surface preparation technology plays a crucial role in maximizing the inherent bonding properties of the adhesive, because its bonding strength is greatly affected by the surface condition. This paper aims to discuss the relationship between the surface condition of metallic materials and adhesion properties. Additionally, it will cover the improvement of adhesion by surface preparation, and efforts to evaluate adhesion strength and durability, while considering the actual environment of use.

Introduction

In the field of metal joining technology, welding and bolted connections play a leading role and are widely utilized across various industries mainly for ferrous materials. On the other hand, for joining parts requiring complex shapes, high dimensional accuracy, dissimilar materials, or areas where heating is to be avoided, adhesive joining takes precedence. Adhesive joining excels in rigidity, thanks to its ability to secure joints over a large surface area, offering high air and water tightness. Moreover, it avoids the distortions and embrittlement caused by heat, often experienced in welding. Additionally, it eliminates the need for pre-drilling and screwing associated with bolted connections, showcasing superior attributes in terms of appearance and productivity. Given these advantages, in recent years, the application of adhesive joining has expanded in the automotive industry for joining dissimilar materials, like joining aluminum and composites, and for improving NVH (Noise, Vibration, Harshness) characteristics¹⁾. Some manufacturers outside Japan utilize adhesive joining in skeleton structures for enhancing the collision safety.

Adhesive joining poses unique challenges compared with bolted connections and welding. When designing the joint strength of adhesive joining, for example, consideration must be given to both adhesion strength and the material strength

of the adhesive itself. From a standpoint of strength stability, it is desirable for the adhesion strength to be higher than the material strength to ensure that any fracture would consistently occur in the adhesive material (hereinafter referred to as “cohesive failure”). Insufficient adhesion strength may lead to delamination at the interface between the adhesive and metal before cohesive failure occurs (referred to as “interfacial delamination”), making it challenging to achieve stable joint strength. Furthermore, since adhesives are typically resin-based, they are prone to undergo changes in state due to water, heat, UV radiation, and stress. Consequently, when selecting adhesives, consideration must be given to their effects in real-use environments²⁾.

Due to these challenges, adhesive joining falls short of providing sufficient strength reliability compared with other joining technologies. Its adoption is limited in applications requiring prolonged use in harsh environments or demanding high safety standards. Therefore, this paper aims to contribute to the understanding and improvement of adhesive joining for metal materials. Specifically, it compiles and explains factors that significantly affect adhesion strength, such as the surface condition of materials, surface modification technologies for achieving stable adhesion strength, and case studies on ensuring long-term strength reliability³⁾.

1. Surface conditions of metal materials and adhesiveness

When bonding metal materials, the adhesive comes into contact with their surfaces, making the surface conditions of the material significantly affect the adhesion strength. In most cases, the surfaces of metal materials are covered with natural oxide films. The effect of material compositions and various processes such as rolling, extrusion, cutting, as well as heating, water cooling, and others during manufacturing can lead to excessive growth of the oxide film, structural defects, alterations and corrosion, creating non-uniform conditions. Additionally, organic matter from processing oil, packaging materials, storage environments, etc., may be adsorbed onto the oxide film, significantly impacting the adhesiveness of metal materials⁴⁾

(Fig. 1(a)).

1.1 Surface contamination

In bonding any substance, not limited to metal materials, the first essential step is to eliminate factors hindering the physical contact between the adhesive and the adherend material surfaces. Due to these factors, residues on the surfaces of adherend materials reduce the contact area between the adhesive and the surfaces of the adherend materials, hindering the intended adhesion strength. Therefore, it is necessary to clean the surfaces of adherend materials before adhesion. While particles and oil content adsorbed on material surfaces can be removed to some extent by solvent cleaning and the like, some substances, such as those forming a strong chemical bond with the oxide film, may not easily be removed. Hence, alkaline degreasing agents are commonly used for chemical cleaning⁵⁾. On the other hand, certain adhesives have properties that allow them to absorb and disperse oil content within the resin. These adhesives can exhibit good adhesiveness even when there is some oil on the surfaces of adherend materials and are used in the automotive manufacturing processes⁶⁾.

1.2 Vulnerable Areas

The next important point is the strength of the material surface. As previously mentioned, the surfaces of metal materials are covered with oxide films that change their states due to alloy composition, manufacturing processes, and storage conditions, leading to structurally or chemically brittle areas. These vulnerable areas, known as weak boundary layers (WBL), become fracture initiation points in adhesive joining and can destabilize adhesion strength.

For example, the surfaces of high-strength aluminum alloys containing magnesium are known to have a brittle, magnesium-containing oxide film

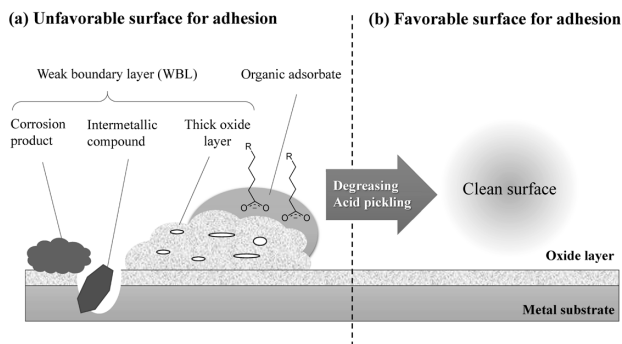


Fig.1 Image of metal surface condition

due to the heat effect during manufacturing, leading to WBL⁷⁾. This oxide film can be dissolved and removed with acid, which is why acid pickling is often carried out as part of the pre-treatment for adhesive bonding of aluminum alloys, typically in conjunction with degreasing processes. After acid pickling, only a thin, well-bonding natural oxide film is present on the surfaces of aluminum alloy, enabling the material's inherent adhesiveness to be expressed (Fig. 1(b)). In addition to acid pickling, processes such as blasting treatment and laser treatment can effectively remove surface contamination and WBL simultaneously, making them useful as pre-treatments for adhesion.

Metal materials vary in manufacturing methods and composition depending on the type, shape, and application of the material. Additionally, manufacturing processes and facilities may differ among manufacturers. Consequently, even for metal materials with the same strength specifications, their surface conditions may not be identical. Therefore, the pre-treatment processes discussed in this section, aimed at removing surface contamination and WBL, are fundamental operations for achieving high strength reproducibility in adhesive bonding.

2. Adhesion mechanism and surface modification of metals

As stated in Section 1, the ability to demonstrate the inherent adhesiveness of an adhesive can be achieved by "resetting" the surface of the metal material by removing contamination and WBL and returning it to a state with only the natural oxide film. Furthermore, a crucial factor is the compatibility between the adhesive and the oxide film. Due to the polymer nature of the adhesive, its chemical structure varies depending on the type of resin composing it. Similarly, the composition and chemical properties of the oxide film vary, depending on the type of base material, leading to differences in adhesiveness based on the combination of adhesive and metal material, as reported in the literature⁸⁾. For instance, Fig. 2 shows the observation results of the fracture morphology after lap-shear tests on epoxy-bonded joints of titanium and copper sheets which had been cleaned and had their surface morphology controlled by blast treatment. In the untreated state, both materials have their metal surface, and interfacial delamination dominates between the adhesive and material. However, blasting treatment changes the fracture morphology to cohesive failure of the adhesive. In the case of copper, the fracture is almost entirely cohesive failure. In contrast, with titanium,

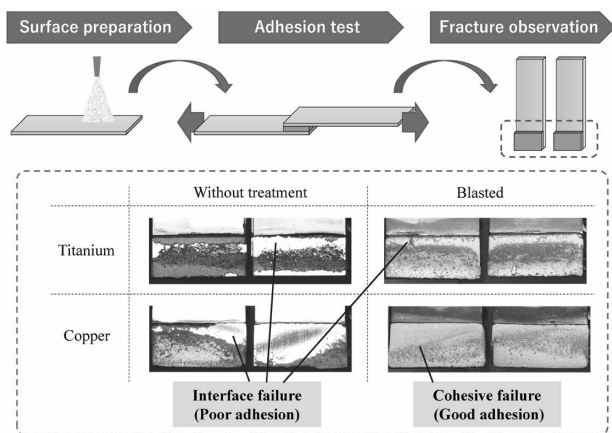


Fig.2 Difference of fracture surface after adhesion test of blast-treated or untreated titanium and copper

interfacial delamination occurs. This indicates that, even when the material's cleaning conditions and surface shapes are identical, there are differences in adhesiveness based on the type of metal material, specifically, the differences in the oxide film.

Moreover, the stability of the oxide film is a crucial factor affecting adhesion strength. While the oxide film immediately after cleaning treatment is immaculate, it easily adsorbs organic matter and moisture from the atmosphere, leading to changes in its surface condition over time, causing a variation in adhesion strength. Therefore, to consistently achieve adhesion strength, a surface modification treatment is necessary to chemically stabilize the oxide film against temperature and humidity changes.

Automotive manufacturers in EU and USA have included surface modification treatment in their standard specifications, in addition to surface cleaning, with the aim of improving the adhesiveness and storage stability of aluminum alloy oxide films. This ensures the long-term stability of adhesive strength⁹⁾. Hence, this section introduces Kobe Steel's research on the mechanism by which adhesive strength manifests at the interface between adhesive and metal material, providing a foundation for understanding the surface conditions of metal materials that excel in adhesion stability.

2.1 Improvement of adhesiveness with silane coupling agent

The adhesion action occurring at the interface between the adhesive and the oxide film has given rise to various hypotheses, including the anchoring effect, chemical bonding such as acid-base bonding and covalent bonds, as well as physical bonding like hydrogen bonding and van der Waals forces. In reality, these factors act in combination, and it is

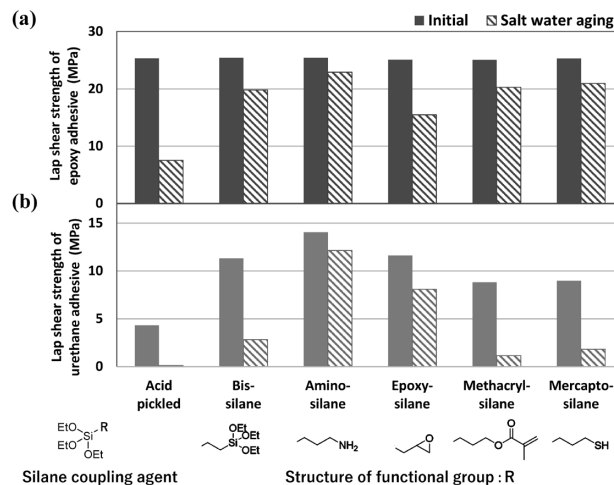


Fig.3 Adhesion test results of silane-treated aluminum alloy with (a) epoxy and (b) urethane adhesive. 6000 series alloy, 1mmt, aging condition; 5%NaCl aq., 40°C, 14 days

believed that their contributions vary, depending on the types of adhesive and oxide film¹⁰⁾. For example, commonly used structural adhesives such as epoxy-based adhesives and urethane-based adhesives are thought to exhibit adhesive strength between the “functional groups of the resin that constitutes the adhesive” and the “functional groups on the surface of the oxide film”^{11), 12)}. Additionally, the improvement of adhesiveness through coupling agents that excel in bonding with both the resin and the oxide film has been reported¹³⁾. Hence, investigations have been conducted into the relationship between the state of the metal surface after surface modification and adhesiveness.

Aluminum alloy sheets surface-treated with silane coupling agents comprising a variety of functional groups have been bonded with epoxy adhesive and urethane adhesive to evaluate the joint strength before and after degradation due to saltwater immersion. The results are shown in Fig. 3. In the case of joints bonded with epoxy adhesive, there is no significant difference in strength before saltwater immersion, as all evaluations have led to base aluminum fracture. However, after saltwater immersion, the use of coupling agents has resulted in less strength reduction compared with the acid-pickled material. On the other hand, in the case of urethane adhesive, the improvement effect of adhesiveness for different types of functional groups is significant. Particularly, the use of coupling agents containing amino groups has demonstrated a superior adhesiveness improvement effect. To elucidate the mechanisms of these effects, the following sections introduce the results of the analysis of the adhesive interface states using

various spectroscopic methods. This analysis has utilized bis-silane and amino-silane treated materials, which demonstrated effects for epoxy adhesive and urethane adhesive, respectively.

2.2 Mechanism of adhesion strength improvement for epoxy adhesive

In recent years, there has been progress in research on the methods of analyzing the adhesion action directly and non-destructively at the interface between the adhesive and the oxide film¹⁴. As an example, Fig. 4 shows the results of the analysis of the electronic state of aluminum oxide film before and after epoxy adhesive coating, using Hard X-ray Photoelectron Spectroscopy (HAXPES). The electron state of aluminum atoms constituting the oxide film is represented by the broad spectrum, and the peak top position shifts to the low-energy side after coating the adhesive (Fig. 4(a)). This indicates the donation of electrons from the functional groups of the adhesive's resin to the aluminum atoms in the oxide film. The electron transfer, namely the acid-base interaction, occurs with the adhesive's resin acting as the electron donor (base) and the oxide film as the electron acceptor (acid)¹⁵. The HAXPES spectrum of the bis-silane treated material shows a greater peak shift after adhesive coating, compared with the acid-pickled material, suggesting a stronger acid-base interaction. Fig. 4(b) presents the analysis results by fitting the HAXPES spectra to the acid-pickled material and bis-silane treated material, separating the peaks corresponding to the chemical bonding that constitutes the oxide film. The analysis has been conducted on the assumption that the peaks labeled Al-O represent the bonding

that makes up the oxide film, the peaks labeled -OH bonds (Al-O-Si-OH, Al-OH) represent the bonding on the oxide film surface, and the peaks labeled -O-C bonds (Al-O-Si-O-C, Al-O-C) represent the bonding with the adhesive. The bonding on the acid-pickled material is Al-OH, while the bonding on the bis-silane treated material includes Al-OH and silanol groups (Si-OH) in Al-O-Si-OH, indicating a higher proportion of OH groups on the oxide film surface. Furthermore, the analysis of the bonding state after adhesive coating has revealed that the bonding quantity with the adhesive is higher for the bis-silane treated material (Al-O-Si-O-C and Al-O-C) compared with the acid-pickled material (Al-O-C). Silane coupling agents are known to generate silanol groups through hydrolysis during surface treatment, and these groups bond with the oxide film (Fig. 5). Silanol groups have a higher acidity than the hydroxyl groups (Al-OH) present

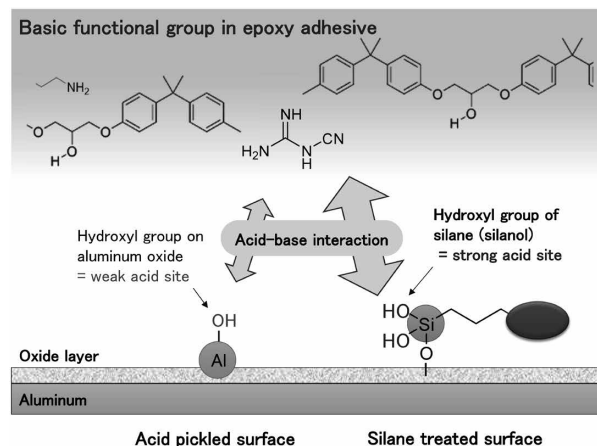


Fig.5 Proposed adhesion mechanism between epoxy adhesive and acid-pickled or silane-treated aluminum surface

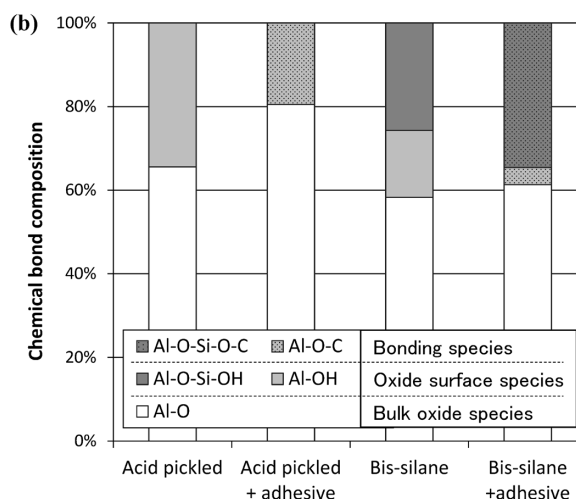
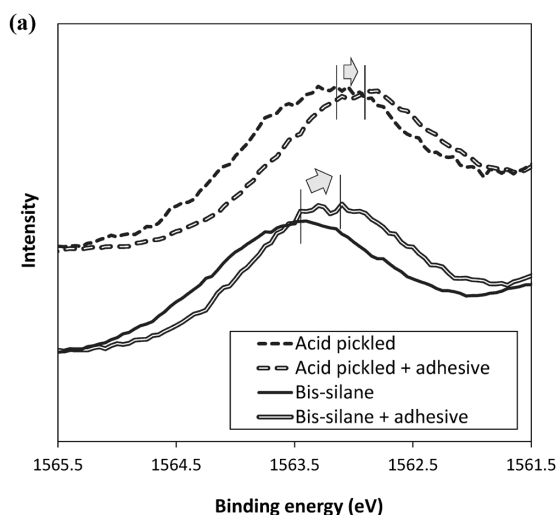
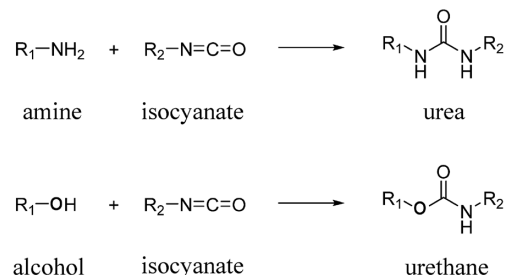


Fig.4 HAXPES results of acid pickled or bis-silane-treated 6000 series aluminum alloy before and after epoxy adhesive coating. (a) Al 1s spectra, (b) curb-fitting result of chemical bonds in oxide layer

in aluminum oxide, and oxides containing silanol groups, such as silicic acid, are known to have excellent compatibility with organic compounds of a basic nature compared with aluminum oxides containing Al-OH^{16), 17)}. Therefore, it is believed that silane coupling treatment results in the generation of acidic silanol groups on the oxide film, leading to a greater and stronger acid-base bonding with the adhesive, improving the bonding properties between the adhesive and the oxide film, and suppressing the reduction in strength due to saltwater immersion.

2.3 Mechanism of improvement of adhesion strength for urethane adhesive

Urethane adhesive is believed to express adhesive strength by the isocyanate (—N=C=O) in the curing agent reacting with the adherend material surface, forming a covalent bond. Therefore, to investigate the change in chemical bonding on the sample surface, isocyanate was coated on the amino-silane treated material, in which improvement in adhesiveness had been observed. The change was analyzed using Infrared Reflection Absorption Spectroscopy (IRRAS) (Fig. 6). In the amino-silane treated material, an absorption peak around 1,640 cm^{-1} is observed. In contrast, the acid-pickled material, which exhibited inferior adhesiveness, shows almost no absorption peak in this region. Therefore, it is believed that this absorption peak indicates the chemical bonding related to the adhesive strength of urethane adhesive. Isocyanate is known to react with various functional groups. For example, it reacts with amines to produce



Scheme 1 Reaction mechanism isocyanate with functional groups

urea (—NHCONH—) and bonds with hydroxyl groups, such as those in alcohols, to form urethane (—OCONH—) (Scheme 1). These reactions are characterized by absorption peaks around 1,650 cm^{-1} . Therefore, the absorption peak around 1,640 cm^{-1} in Fig. 6 is attributed to the urea bonding generated by the reaction between the amino group introduced to the oxide film surface through amino-silane treatment and the isocyanate in the urethane adhesive. The covalent bond formation between the oxide film and the adhesive has improved adhesiveness. Furthermore, with other silane treated or acid pickled materials, varying degrees of adhesion strength have been obtained. It is assumed that these agents introduced silanol groups onto the oxide film surface through silane coupling agents or that the hydroxyl groups on the oxide film surface bonded with the isocyanate, generating urethane bonding, and expressing adhesion strength. Urethane adhesive typically exhibits limited adhesiveness to metal materials and usually requires the use of specialized primers or coatings for pretreatment. However, performing modification treatment on the oxide film as described provides a certain level of strength stability without the need for a primer.

2.4 Effect on dissimilar metal materials

As observed in the improving of aluminum oxide film's adhesiveness to the adhesive by modifying the film, comparable effects can be anticipated for dissimilar metal materials. Various metal materials were subjected to blasting treatment followed by silane coupling treatment, and the adhesiveness to epoxy adhesive was evaluated as shown in Fig. 7. It is observed that, in addition to aluminum and stainless steel, the adhesiveness of titanium, which exhibited interfacial delamination with blasting treatment alone, as demonstrated in Fig. 2, can also be improved. By enhancing the adhesiveness of metal materials through appropriate surface treatment and suppressing interfacial delamination,

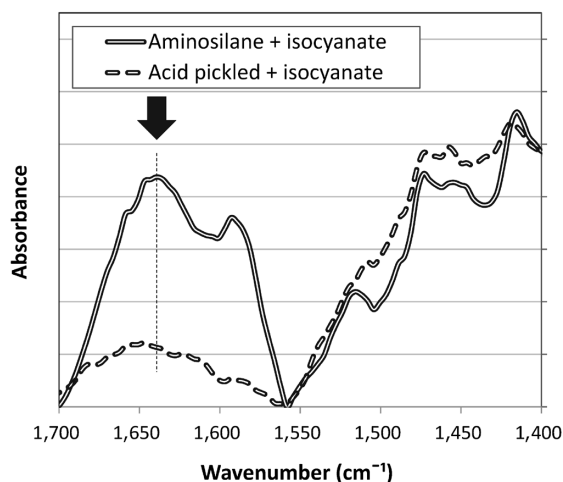


Fig.6 FT-IR spectra of acid-pickled and amino-silane-treated 6000 series aluminum alloy after isocyanate compound coating (data was taken by reflection absorption spectroscopy method under nitrogen stream)

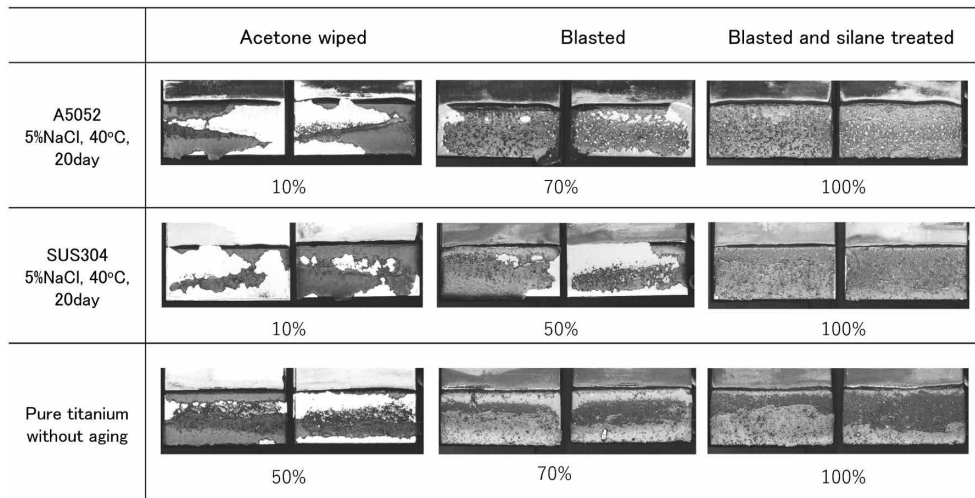


Fig.7 Fracture surface structure and rate of cohesive failure mode of metals with different surface preparation

it is possible to mitigate the decrease in strength in adhesive joints, thereby enhancing the reliability of long-term strength. Furthermore, the establishment of suitable surface modification technologies for different materials and adhesives holds the potential for further utilization of adhesive joining, especially in the context of dissimilar material joints.

3. Reliability of adhesion strength

When using adhesive bonding over an extended period, it is crucial that the designed joint strength does not easily deteriorate in the face of external environmental changes, such as temperature, humidity, ultraviolet radiation, and repetitive stress. In other words, adhesive durability becomes paramount. The durability of adhesive itself can be estimated to some extent by controlling the type of resin and the skeleton structure that constitutes the adhesive and evaluating the resin material in practical terms. On the other hand, predicting the durability of adhesion strength with the material involves considering changes in the adhesive, oxide film, and the interface between them, making it challenging. Hence, this section will introduce the mechanisms through which strength reduction occurs at the adhesive interface due to water, which requires particular attention when using adhesive bonding over an extended period. Also discussed are approaches to reliability evaluation that take these effects into account.

3.1 Factors of strength reduction

Adhesive undergoes changes in its chemical structure during the curing process, often resulting in volume changes such as expansion or contraction,

as well as the generation of bubbles. Particularly at the adhesive interface, structural defects and compositional imbalances are prone to occur. As a result, the interface between the adhesive and the metal is more susceptible to water infiltration than the adhesive itself¹⁸⁾. When water infiltrates the adhesive interface, it not only hinders the adhesion action between the adhesive and the oxide film, but also leads to various changes in the state, such as hydration of the oxide film and alterations in pH, as predicted by theoretical calculations. Consequently, this can result in a reduction in adhesion strength¹⁹⁾.

Therefore, water infiltration into the adhesive interface and the associated changes in the oxide film's state are significant factors causing a reduction in the adhesion strength of metal materials. The impact of these phenomena becomes more pronounced in the presence of factors that accelerate these processes. For instance, higher environmental temperatures cause the resin to become plasticized, accelerating the diffusion of water and, consequently, promoting strength reduction. Additionally, applying stress to the joint exacerbates the reduction in strength²⁰⁾. It is known that applying stress to the joint has the effect of expanding voids in the resin or adhesive interface, facilitating water absorption. Furthermore, the stability of the metal oxide film significantly decreases in the presence of chloride ions, leading to corrosion reactions, dissolution of the base metal, and changes in pH that can invade the adhesive interface. Therefore, special attention is required when using adhesives in environments exposed to saltwater (Fig. 8)²¹⁾.

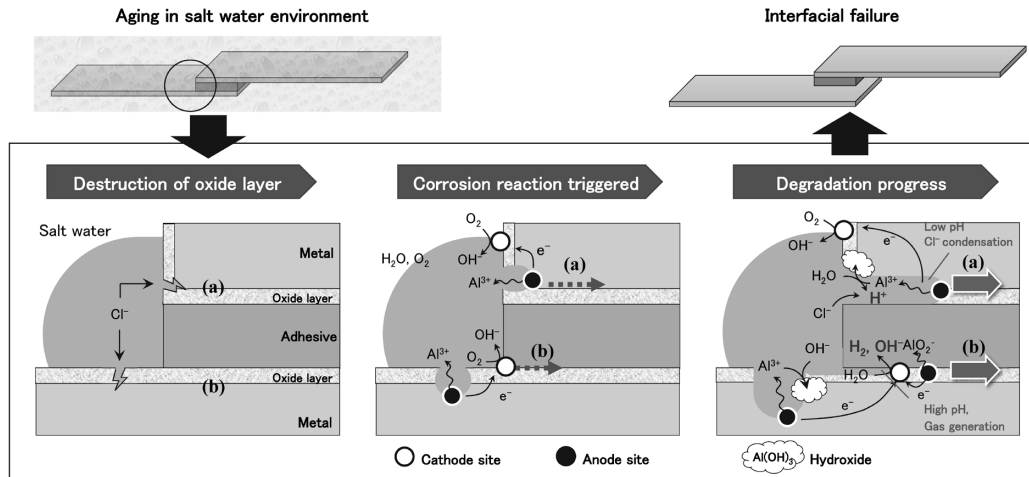


Fig.8 Degradation mechanism between adhesive-oxide layer interface in salt water condition
(a) crevice corrosion, (b) cathodic delamination

3.2 Measures to inhibit strength reduction

To mitigate the reduction in adhesive strength, two key measures are crucial: “preventing water infiltration into the adhesive interface” and “restraining changes in the oxide film’s state.” Regarding measures on the metal side, the surface modification technology discussed in Section 2 becomes essential.

To prevent the infiltration of water into the adhesive interface, it is crucial to enhance the adhesion between the adhesive and the oxide film, eliminating crevices and voids between them, and thereby preventing the creation of pathways for water infiltration. As illustrated in Fig. 3, the use of a silane coupling agent has been shown to increase the bonding between the adhesive resin and the oxide film, contributing to improved adhesion durability in a saline environment. Furthermore, as a modification treatment to suppress changes in the oxide film’s state, various protective film treatments have been employed for surface pretreatment in painting, such as chromate treatment, anodizing, and zinc phosphate treatment. These treatments create stable protective films against acids, bases, and chloride ions and have been utilized for an extended period²²⁾. On the other hand, while these protective films excel in chemical stability, the protective film itself may become a starting point for fractures depending on the adhesive’s strength characteristics. Hence, the selection of an appropriate film depends on the adhesive used, the operating environment, and the required product lifespan.

3.3 Strength durability in real environment

When designing a product using adhesive joining, it is essential to consider the expected decrease in adhesion strength over the product’s lifespan and to design joint strength accordingly. To achieve this, it is desirable to conduct an accelerated evaluation that intentionally exposes the joint over a specific period to environmental factors known to have a significant effect on adhesion strength, as described in Section 3.1. This evaluation allows for predicting the strength lifespan in actual market conditions.

The following presents examples of efforts to improve adhesion strength reliability in the automotive market outside Japan. The auto manufacturers in the EU and USA have established their own criteria. In Europe, as part of accelerated tests simulating market environments, adhesive joints undergo cycle tests combining saltwater spray and drying for several thousand hours, with criteria specifying that interfacial delamination after strength testing remains below a certain percentage. In North America, a different approach is taken, where pre-oiled test pieces are adhered, subjected to environmental cycle tests with applied stress, and evaluated for their strength lifespan²³⁾. To meet these stringent testing criteria, surface modification treatments applied to aluminum alloys include barrier treatment using a composite oxide with titanium and zirconium (TiZr treatment) in Europe and a coupling agent treatment (A951 treatment) based on organic phosphoric acid in North America^{9), 24)}. These treatments have been adapted to volume production. Both treatments have demonstrated the ability to maintain the majority of the initial adhesion strength even after more

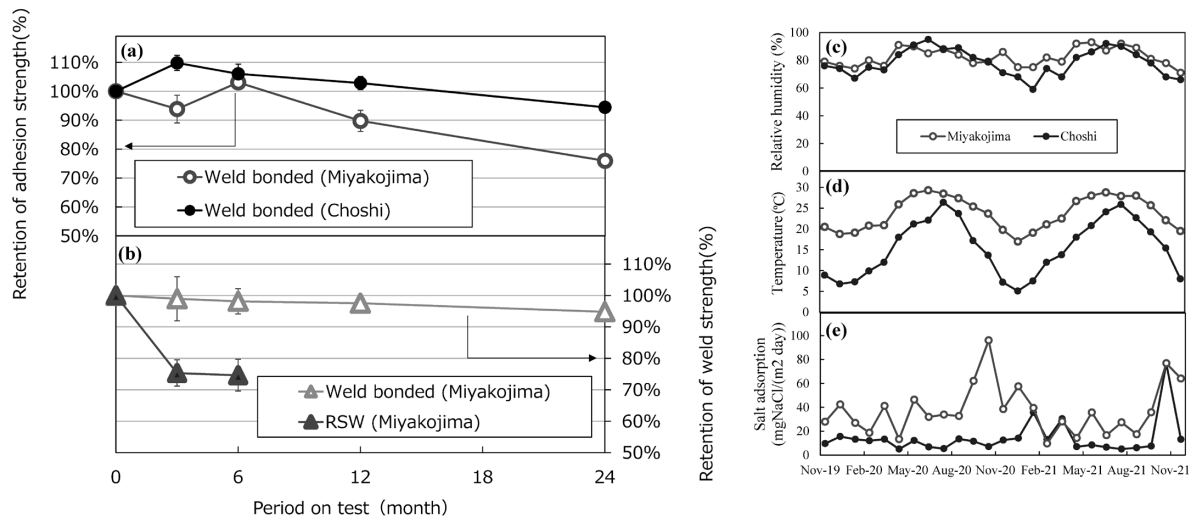


Fig.9 Joint strength result of bonded steel under climate exposure test (a) weld-bonded specimen at Miyakojima and Choshi, (b) weld-bonded and spot-welded specimen at Miyakojima, climate conditions during exposure period (c) relative humidity, (d) temperature, (e) salt adsorption

than ten years of use in various regional market environments. It is inferred that reliability has been established over an extended period on the basis of the results of “1 to 2 months of accelerated laboratory testing” and “several years of actual vehicle driving tests”²⁵⁾.

Also in Japan, there is a correlation being established between the residual strength of adhesive joints evaluated by recovering vehicles that have been on the market for several years and the accelerated laboratory evaluation results based on the adhesive used in those vehicles. These efforts have led to estimating a correlation between “short-term accelerated tests in high-temperature and high-humidity environments” and “results equivalent to several years of actual driving”^{26), 27)}.

Finally, the benefits of using adhesive in combination with other joining methods for durability are introduced. Steel sheets were joined using resistance spot welding or a weld-bonding method combining adhesive and resistance spot welding. After exposing them to environmental conditions for a certain period at the outdoor testing sites in Choshi City or Miyakojima City, the remaining strength of the joints was evaluated, and the results are shown in Fig. 9²⁸⁾. As shown in Fig. 9(a), even when exposed to harsh corrosive conditions such as the sea breeze and sunlight for two years, the majority of the initial strength is maintained. This indicates that the adhesive used in this test, a structural epoxy adhesive for automobiles, possesses high durability. Furthermore, in Miyakojima City, where the average temperature is higher and the salt adhesion level is greater, a more significant decrease in strength is observed

(Fig. 9(c)-(e)). This result reflects the influence of the factors causing strength reduction as discussed in Section 3.2. It should be noted that in joints joined only by spot welding, corrosion occurs around the welded area, and the joining strength decreases shortly after the exposure. In contrast, the weld-bonded joint whose welded area is sealed with adhesive is protected from the corrosive environment, resulting in the suppression of strength reduction in the welded area (Fig. 9(b)). Thus, by combining adhesives with other joining technologies, synergistic effects can be expected, leading to an enhancement of joint strength over an extended lifespan.

Conclusions

The expectations for adhesive joining are increasing year by year. In the automotive industry, particularly in response to the electrification trend leading to the lightweighting of vehicles and increased production of batteries, the adoption of adhesive joining is expected to expand further. Additionally, in non-automotive industries, there is a growing demand for adhesive joining of dissimilar materials and productivity improvement. Considering societal challenges such as a shortage of welders²⁹⁾, adhesive joining is anticipated to become one of the primary joining technologies for metal materials across various industrial sectors.

On the other hand, as mentioned in this paper, to be able to rely on adhesive joining, it is crucial to consider the surface conditions of metal materials. Proper management of their state, including surface treatment, becomes essential. Furthermore, since

the products, environments, and performance requirements vary among users, it is important to organize the relationships among adhesives, metal materials, their surface conditions, and the strength durability in the specifications in each case. With the consolidation of such cases and experiences, there is an expectation that the strength reliability of adhesive joining will continue to improve.

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