

<Review>

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Connecting Technology with UV-High Performance and UV-Curable PSA in Electrical/Electronic Industry

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1. Introduction

Industry is changing rapidly. Various techniques have been fused, and the fusion is made to the birth of the new technology. PSA is placed in the center of such changes as shown in **Fig. 1**. It has been a core material in the industry. PSA has various advantages in terms of speed of process, friendly approach to the environment and the new function. PSA is a typical fusion

as the initiator after an exposure to UV light.

In general, UV curing technology can be divided into UV polymerization technique and UV curing technology. UV polymerization technique is similar to polymer technology to prepare general linear polymers; its process has the advantage that it can be easily applied for the bulky polymerization. UV curing technology is to form three-dimensional polymer structures by cross-linked multifunctional monomers such as HDDA (1, 6-

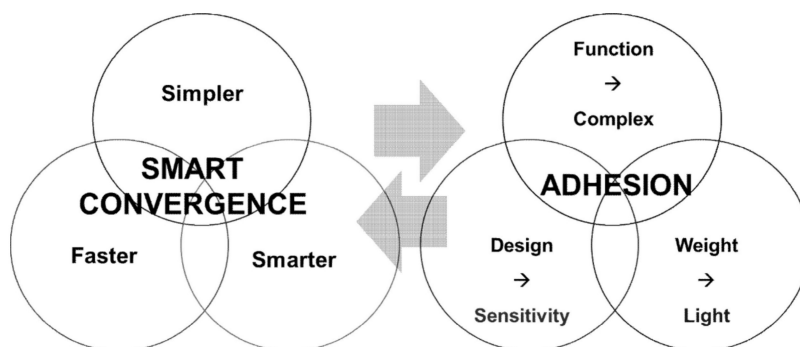


Fig. 1 Keyword of Smart Convergence and Adhesion

technique with several fields, and the application of PSA that is fused with UV technique among them is extended rapidly.

2. Conventional Approach of UV Curing Technology

2.1 Review of UV Curing Technology

The UV curing technology is a technique to form polymer structures by chain reaction for ring structures or double bonds by activating radicals or cations

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hexandiol diacrylate), DPHA (dipentaerithritol hexacrylate) by UV initiation.

Advantage of UV curing
Solventless process: non VOCs
Room temperature processing
Fast curing system: low energy consumption
High speed process
Small handling space
Excellent finishing processing

Because UV-curable resins are cured instantaneously when irradiated with UV, it is used in a key technology in the field of adhesive. It has useful features that are little changing of volume after curing (compare thermal curing), transparency and fast drying, so it is often used in the adhesion of precision mechanical parts and optical components.

2.2 IPN structure of PSA

IPNs, Inter-penetrating polymer networks are comprised of the blend of two or more numbers of polymer components. At least one of the components has internal bridge — crosslinking— and which makes network structures to form a permanent entanglement structure¹⁾. There are two methods to build a IPNs structure, sequential and simultaneous process. The former one, sequential methods constructs a network and polymerize the sequent crosslinking system. The latter one, simultaneous method makes two network components being polymerized simultaneously to form a IPNs structure. At the initial stage of development of IPNs structure, the step polymerization induced by thermal radical or cationic radical method was performed to form IPNs structure²⁾. Nowadays, however, the radical polymerization induced by photochemical method would be preferred because of its various advantages against the conventional method; eco-friendly polymerization without solvent, no need of additional thermal process in owing to room-temperature polymerization³⁾, high process ability. Along with this, the photo-crosslinking process forms IPNs structure rapidly preventing the phase separation which can be occurred by the conventional thermal polymerization process on account of mixing the two different network system with low miscibility⁴⁾.

For these various advantages, IPNs structure in-

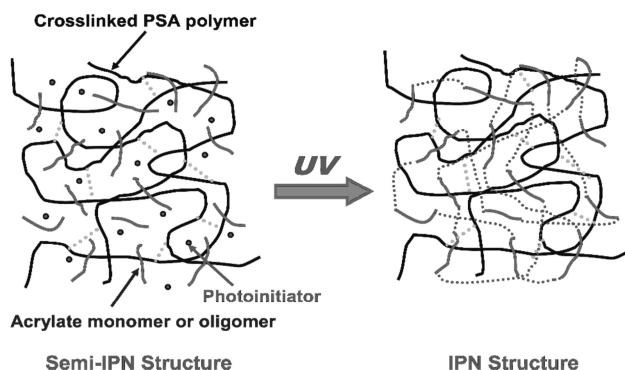


Fig. 2 IPN structure induced by UV-crosslinking.

duced by UV-crosslinking has been made the most of pressure-sensitive adhesives as well as many other applications. The sequential method to form IPNs structure by photochemical way is shown in Fig. 2. It shows the process of making the pressure-sensitive adhesive with IPNs structure by UV irradiation to the photoinitiator which releases the radicals to polymerize the acrylate monomers or oligomers with cross-linked PSA polymers. Our research group has studied about the mechanical and chemical properties of the PSA with IPNs structure which was synthesized by UV-irradiating to the semi-IPNs system of the PSA polymer and multifunctional monomer⁵⁾.

Our research group reported on the crosslinking performance of the IPN system composed of acrylic binders, tri-functional monomers and a photo-initiator⁶⁾. The crosslinking performance and reaction rate were dependent on the types of the multifunctional monomers, trimethylolpropane triacrylate (TMPTA) and trimethylolpropane ethoxylated triacrylate (TMPEOTA) which have different molecular weight and structure with the double bond in their branch to form a crosslinking by UV-irradiation-induced radical reaction. These multifunctional acrylates accelerate the crosslinking of the system by radical polymerization. When the crosslinking between the binders and multifunctional monomers is formed, it could be lead to the IPN structure which gives the PSAs many advantages, like chemical resistance, hardness and scratch resistance.

In order to characterize the kinetics of the IPN forming process by UV irradiation, photo differential scanning calorimeter (photo-DSC) is chosen to monitoring the exothermic reaction from photo-polymerization. Evaluating the rate of polymerization and identifying the reaction kinetics were performed by analyzing the

profiles of heat of reaction versus time, provided by photo-DSC. Photo-DSC thermograms of the two tri-functional monomers, TMPTA and TMPEOTA blended with a photo-initiator shows a comparison of the rate of crosslinking reactions by TMPTA and TMPEOTA, respectively. TMPTA, which has a shorter chain length in its structure, is to be more reactive at photo-crosslinking on account of the lower concentration of double bonds in the structure and the enhanced mobility to increase the crosslinking density within the free volume polymer blend system. Hence the reaction of the crosslinking is predicted to be performed much rapidly in TMPTA-based PSAs than in TMPEOTA-based PSAs and it correspond with the results of the photo-DSC as shown in Fig. 3.

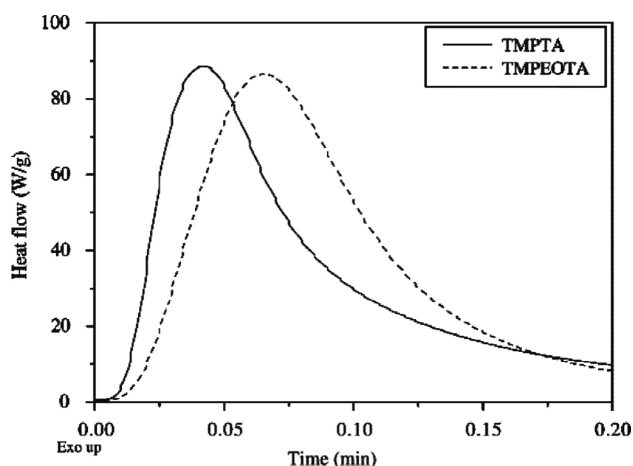


Fig. 3 Heat flows of the tri-functional monomers measured by photo-DSC.

Our group also studied the crosslinking behaviors, PSA performance and the thermal resistance of IPN structured PSAs with different types and contents of multifunctional monomers as function of UV dose. Gel fraction stands for the value of the insoluble fractions of cross-linked or network polymers. As the crosslinking process goes, linear binder in organic solvents turned into semi-IPN structured PSAs by radical polymerization through the reaction of the double bond of tri-functional monomers. After the UV irradiation, the gel fractions of the three blends increased up to 80–90% which suggests the compact IPN structured PSA is formed using TMPTA shown in Fig. 4 (a). Since the TMPTA has a shorter chain length which gives it a mobility to make a much dense crosslinking structure within the system compared to the result of the gel fraction of TMPEOTA as shown in Fig. 4 (b).

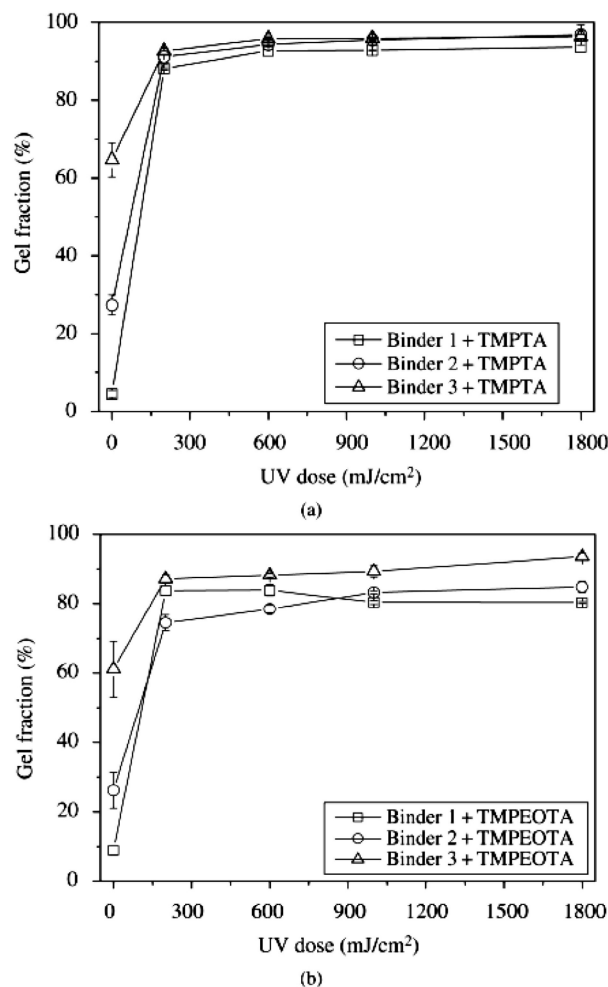


Fig. 4 Gel fraction of the semi-IPN structured PSA with the blend of binders and multifunctional monomers.

In general, short chain length monomers react much rapidly which leaves many monomers unreacted in the cross-linked structure of PSA which lowers the rate of conversion and effects the entanglement of multifunctional monomers on binder miscibility and finally decide the ability to form an IPN structure of the PSA⁷⁾.

2.3 UV-Cross linkable Hot-Melt PSAs

UV-cross linkable Hot-melt pressure-sensitive adhesives (HMPSAs) have been developed to overcome its weaknesses in heat stability and chemical resistance which are originated from its composition, usually containing styrene-isoprene-styrene (SIS) or styrene-butadiene-styrene (SBS) block copolymers, tackifier, plasticizer and other additives. In general, the cohesion of HMPSAs are obtained by the crosslinking structure of the styrene based end-blocks. Since this temporary physical network, however, can be easily disassembled at the temperatures of hot melting environment. The

application of SBS-based PSA is limited. Hence the permanent chemical crosslinking reaction induced by UV irradiation would be needed for its adhesion properties and heat resistance in accordance with the proper selections of photo-initiators and crosslinking agents⁸⁾.

2.3.1 Crosslinking system of HMPSAs using the thiol-ene reaction

UV-crosslinkable HMPSAs can be formed by the thiol-ene reaction to invest better heat resistance as well as high adhesion properties. Chemical crosslinking was achieved by the incorporation of photo-induced thiol radicals with the double bonds of the polymer branches.

In such case of incorporating the UV-crosslinking agent into the system, the step addition reaction of the sulfenyl radical ($\cdot\text{RS}$) with chain transfer process occurred and induce an intermolecular polymerization which makes permanent chemical crosslinking structure as shown in Fig. 5⁹⁾.

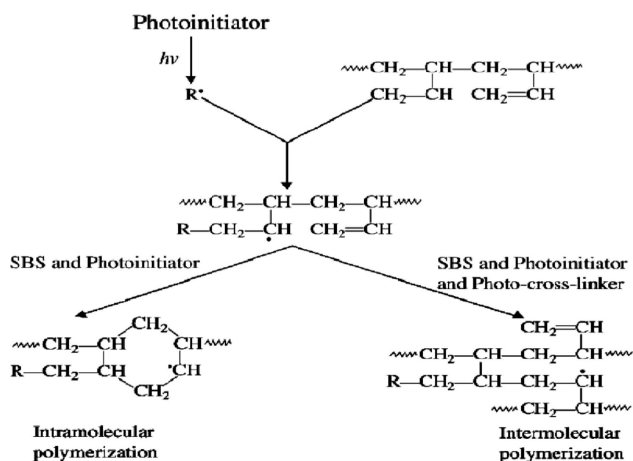


Fig. 5 Chemical crosslinking of the HMPSAs by the thiol-ene reaction⁹⁾.

2.3.2 UV-crosslinked system and the PSA performances

Our research group has studied optimizing the blending condition which has vinyl groups to form a crosslinking structure by UV-irradiation¹⁰⁾. The mechanical and thermal properties of a crosslinking system of HMPSAs are controlled through the combination of types among various photo-initiators and crosslinking agent. We have compared two types of photo-initiators being used in HMPSAs system, each with having different thermal stability. In order to achieve effective UV-crosslinking system, the photo-initiator and the crosslinking agent which have a relatively high heat stability are essentially required.

Comparison of the adhesion properties of HMPSAs such as probe tack, peel strength and Shear adhesion failure temperature (SAFT) test values was performed between the two HMPSAs using Irgacure 819 of high thermal properties and Micure BK-6 of poor thermal properties, respectively.

The effect of the UV crosslinking system of HMPSAs with different degree of UV radiation appears distinctly as in the three values of PSA performances shown in Fig. 6. The probe tack value appears inert as the function of UV-dose as both in two systems using the different photo-initiators. On the other hand, the value of peel strength and SAFT are changed in accordance with increasing of UV dose. As start with the concept of the peel strength, it means the sum of the energy required to break the bonds between a PSA and a substrate in peeling process. Since its own elongation property goes down with UV radiation, peel strength of UV-cross-linked HMPSAs is simultaneously reduced. This phenomenon can be prevented by incorporating the small molecules like tackifier into the UV crosslinking system to enhance the mobility of the HMPSAs so that they could be sustained onto the sub-

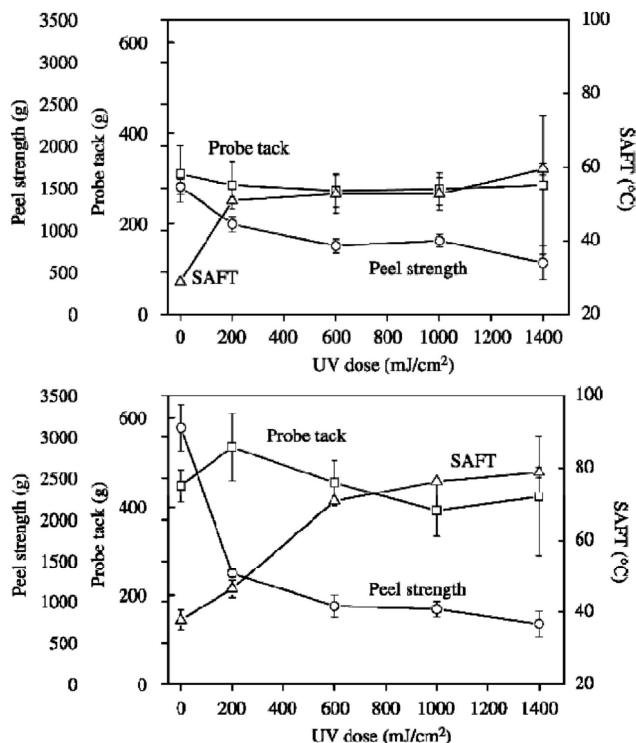


Fig. 6 The effect of the UV irradiation with different degree of UV dose on the probe tack, peel strength and SAFT performances; UV-crosslinked HMPSAs with Irgacure 819 (top), UV-crosslinked HMPSAs with Micure BK-6 (bottom).

strate even after the UV crosslinking procedure.

The UV-cross-linked HMPSAs with including the photo-initiator of high heat stability show high cohesive strength and high heat resistance figured by SAFT results according to the UV dose. In general, SIS- and SBS-based PSAs, in this case HMPSAs, easily lose the cohesive property above their glass transition temperature (T_g), which is relatively low for their application temperature. This SIS- and SBS-based PSAs became disentangled above the T_g and their styrene block structure become broken down to lose the cohesive strength within the system¹¹⁾. The permanent chemical crosslinking induced by UV irradiation gives sustainability to the system, without collapsing the structure, and provides high heat resistance and chemical resistance which were required in the field of the application of HMPSAs.

3. UV Curing Technology of Electric-Electronic Industry

3.1 Adhesives of Electric-Electronic Industry

The electric and electronics industry can be the center point of modern industries experiencing the most rapid change. Looking at the characteristics of electric and electronic parts, the parts need specialized materials and process technology for each part, the parts not only have a very short life cycle and the development period is short but time it is necessary for excellent quality and low price at the same.

Development of parts are continued for downsizing, high speed/large capacity and multifunctional parts. And new materials and technology are steadily applied. Particularly, the using of adhesive materials is expanding, its utilization has been maximized not only in the procedure for utilizing parts but also at the stage of manufacturing and managing parts. Recently, the direction of technical development of adhesive materials required in these industry changes is as follows.

- 1) Multifunctionalization
- 2) Simplification of process and material
- 3) Compatible with various forms of materials
- 4) Cooperation with new materials and new equipment
- 5) High airtightness and high reliability
- 6) Excellent rework characteristics
- 7) Minimum contaminants

3.2 UV Technology of Semi-Conductor Industries

3.2.1 DAF for Semi-Conductor Handling

The DAM (Die Attachment Material) of the packaging material in the semiconductor field has been used since the 1980's Dual Inline Packages (DIP) system was developed. Initially, EMC (Epoxy Molding Compounds) material led QFP (Quad Flat Packages) and TSOP (Thin Small Outline Packages) market.

In the early 1990's, BGA (Ball Grid Array) was developed for systems with fine pitch, and the use of Film type DAM increased. DAM means a description that is utilized for bonding a substance to a silicon chip. This process has been evaluated in a very important process because it is the most important part of determining the reliability and circuit performance during the integration process of IC chips.

DAF (Die Attached Film) is a composite film made by mixing silver particles with thermoplastic polymer material, which was first introduced by Dupont in 1988. Initial DAF was used only for fastening materials it was difficult to solder the inside with conductivity. However, Nitto Denko introduce a new concept of DAF that can integrate the dicing process and the Die Attachment process and can be the founder of the current integrated DAF.

The key to the utilization of DAF is performance control of adhesive strength through UV curing. There are performance control studies on adhesives utilizing multifunctional monomers. The adhesive strength was formed to be close to 0 through UV curing, and it was designed so that it can be de-bonding easily. Application of these technologies can reduce the energy consumption generated in the process and ensure a high processing rate at the same time

3.2.2 Temporary Bonding-Debonding Adhesives for 3D packaging

The semiconductor industry is evaluated in industrial fields that develop so rapidly that they are called flowers in the electric and electronic industry. Moore's Law and Huang's Law are presented, and annually new performance and new size chip (Chip) have been introduced. In these semiconductor changes, recently the most important key words are Thin Wafer and 3D Stacking. Research has been actively pursued to adopt a three-dimensional structure in order to design and manufacture semiconductors to overcome the limitation of in existing 2D regions that produce more

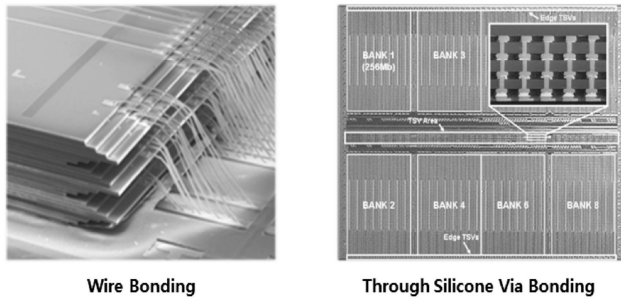


Fig. 7 MCP structure with wire and TSV.

highly integrated materials. The stacked system of these semiconductors is called MCP (Multi Chip Packaging). MCP packaging technology began to appear in the latter half of the 1990s, but at the early stage there was a limit to the manufacturing structure as packaging progressed through wire¹²⁾.

Recently, TSV (Through Silicon Via) construction method has been proposed to eliminate these wires and increase integration rate more as shown in Fig. 7. In TSV, vertical stacking technology opens a fine hole (Via) through thin wafer, and forms an electrode penetrating the chip by being filled with a conductor, and connect to the upper chip and the lower chip by the hole.

The reason for the change of the MCP paradigm is that packaging until the mid-1990s was important to provide the physical protection of the chip and the electrical connection between the board and the chip. The cause of the change in the MCP paradigm is that it was an important issue for the package to provide physical protection of the chip and electrical connection between the board and the chip until the mid-1990s, however, the role of packages is becoming increasingly important in aspects of multi-functionality, optimization of performance, resolution for problems of heat generation as well as high-performance of chips and highly integrated packaging technology is expanding according to technological trends requiring the integration of many semiconductors on a limited board recently. This means that the size of memory chip and other chip would also increase as additional functions are added up to the mobile device. As a result, the importance of packaging technology for making plural chips to a single product or downsizing is increasing.

In order to respond to the needs to miniaturization and high performance, the development of packaging technology has advanced rapidly with that of

semiconductor processing technology. The package manufacturing technology as a post-process of conventional semiconductor processing technology is a next-generation promising industry in Korea which is leading semiconductor technology and the market size and its demand tend to increase sharply. Therefore, the research for temporary bonding-debonding (TBDB) adhesives as its core technology is also considered as the essential.

The reason why the development of temporary bonding-debonding (TBDB) adhesives is important is that a simple design for materials has the limit to overcome the feature that physical properties required for bonding and debonding steps are conflicting. The adhesives have a special characteristic that they should show an excellent high-durability in terms of a strong bonding force, an ultra-high heat resistance, non-outgassing and low TTV in the bonding process while present the properties as low bonding materials that have no-damage, no-residue and a free debonding force in the debonding process¹³⁾. Since the utilization of thin wafer has extended and is accompanied by an important issue of protection of wafer, the top priority problem to resolve in TSV method in the current especially,

3.3 UV Technology for Convergence Technology and Mobile Industry

Recently, the expansion of the use of mobile devices and diversification of the special functions, multiple systems are now being established in one equipment. Mobile device is integrated in the chip-set change essential techniques with performing a varieties of a functions as in for display, communications, applications and so on. Since each of the systems would work with independent process, Integration of the chipset is another factor leading to interference between each other and lead to an abnormal operation of the circuit¹⁴⁾. In particular, display distortion, noise and radio interference caused from typical drive of electromagnetic interference issues, serve as a deadly problem to the conventional mobile devices. Recently, electromagnetic interference shielding technique is supplied in the form of a film and pressure sensitive adhesive (PSA) is commonly used to attach this new film-based system.

3.3.1 Electrically Conductive Adhesive

Electromagnetic shielding is wrapped around a

certain part of the area of a conductive refers to the inside so that the electromagnetic fields generated inside is not affected from the external electromagnetic fields or vice versa, below the outside. For cutting off the electromagnetic field permeating the material to the external, reflecting or absorbing mechanism should be secured within the material system by incorporating the filler, so called, shielding material which has electromagnetic shielding property mainly induced from the electrical conductivity of its own.

Electrically conductive adhesive (ECA) refers to an adhesive which has electrical conductivity enabled by incorporation of a conductive component that comprise over 70% of the total mass of an electrically conductive adhesive. The conductive component, filler particles, usually suspended and in polymer matrix with higher content making densely packed agglomeration¹⁵⁾. This dense or linked structure of the conductive components makes a conductive path which connects the two separated section of the system through the polymer matrix and in this way makes electric current within the system possible as shown in Fig. 8. Due to its main benefits of the circuit assembly and the electromagnetic shielding, ECA has been widely introduced in vari-

ous state-of-the-art industries.

There are many different kinds of shielding material, but most are metals, used in the electromagnetic shielding effects of materials to be used. Specific electric conductance and relative permeability can be largely changed in accordance with the kind of metal which determines the shield performance accordingly. Through the numerous research, Silver series of electromagnetic shielding material is going on the research full-speed ahead even for nowadays¹⁶⁾.

3.3.2 Multiple film layer structure and PSA for the display system

Generally, the display system for the electrical equipment, like mobile devices, is made up of multilayered structure of the multifunctional film with a number of optical films. This multilayered structure would face the contraction and distortion from the external environment of daily using. Under this consecutive stress and strain, the pressure sensitive adhesive becomes strongly attached to the rigid body of the structure, minimizing a fault within the whole sections of the product. On the other hand, in case like flexible display, it does not include the rigid body inside of the system which makes it hard to prevent the deformation phenomenon. To sum up, the multilayered of the films with each having different function of its own would be needed in order to secure the mechanical and functional stability of the product.

Our research group suggested the multilayered film structure for the adhesion of the display of smart devices with five layer as shown in Fig. 9 having much thinner and compact structure compared to the conventional one. The top layer, multifunction layer forms adhesion to the circuit board and serves as the stress relief layer which solves the flexural stress, tensile

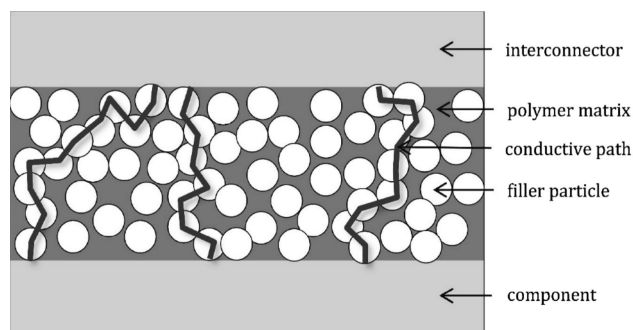


Fig. 8 The internal structure of electrically conductive adhesive

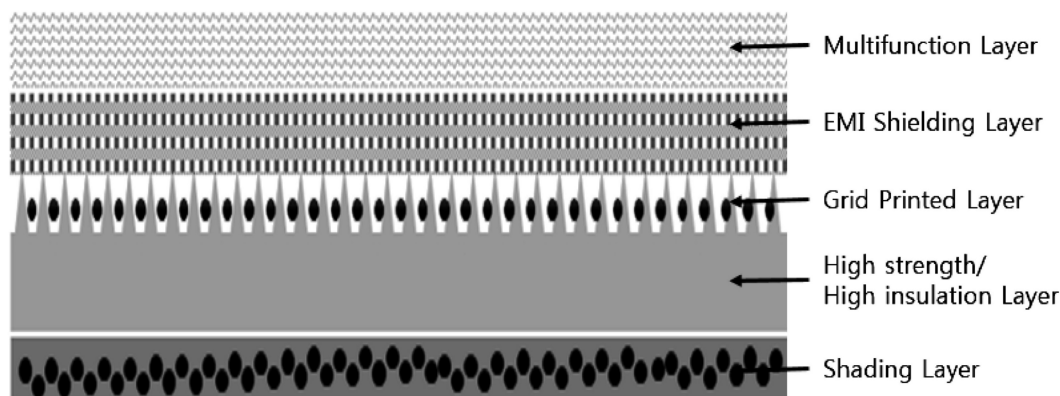


Fig. 9 The internal structure of electrically conductive adhesive

stress, contraction issue and CTE matching arising from the display system. The second layer of the structure is comprised of a thin EMI shielding layer and flexible EMI shielding layer. The thin EMI shielding layer shows major performance of the shielding of the film. For reliable positioning of the conductive components, this layer should be implemented within $10\mu\text{m}$ and additional protective layer is required in case of external exposure of the conductive fillers. The flexible EMI shielding layer serves as the protective layer as to prevent oxidation of the thin EMI shielding layer and is comprised of high conductive polymer materials via the hybrid effect with thin EMI shield layer securing the reinforcement in the overall film performance.

Next, the high strength/high insulation film layer determines the strength and mechanical stability of the whole system and prevents the shot problem occur in circuit boards. The final layer of the film structure is a shading layer. In accordance with the development of flexibility of OLED, front visibility gains recognition to be very important. The shading layer blocks the light from the rear and prevents the leaking of the light in front. It also contains an organic binder which can prevent the oxidation of the nano-imprinted EMI shielding layer.

3.3.3 Conductive filler within the PSA curing system

The performance of conductivity of the electrically conductive adhesive (ECA) appears differently according to the type and shape of the conductive filler used; isotropic conductivity or anisotropic conductivity. Especially, an isotropic conductive adhesives show sheet resistance along the polymer resin, therefore, they are to be used as EMI shielding material which is our very target of the research. Our research group studied the optimal conditions of using ECA as in for their pur-

poses, electrical conductivity and adhesion, using silver particles as the conductive filler within the polymer matrix system. Since these conductive components such as silver particles are hard to be homogeneously distributed within the viscous polymer resin because their high content and the high specific gravity makes them precipitated and agglomerated, it needs to be analyzed in accordance with the type of fillers under optimized dispersion condition.

There are many type of silver particles each having different shape and corresponding aspect ratio. We have chosen two types of silver particles, spherical-shaped type and flake-shaped type whose electrical, adhesion properties and curing behavior show clear comparison for the research.

Flake-shaped filler has a large plate morphology which gives it two trade-off characteristics. Although this plate morphology of flake shaped filler grants anisotropic conductivity to the ECA functioning as EMI shielding material, it is hard to be dispersed and makes the polymer resins difficult to be cured by UV irradiation. Its application of PSA to the circuit or panel assembly is hard to be achieved. On the other hand, spherical-shaped filler has a relatively small dimension which makes it easy to be dispersed and curable by UV irradiation. Hence the hybrid system which combines the pros of each filler systems, anisotropic conductivity and UV-curable property, is required. The hybrid filler system was prepared through the mixing of the two different fillers being introduced into the polymer resin and taken by scanning electron microscope shown in **Fig. 10**.

4. Conclusion

Rolls of PSA are become the most important materi-

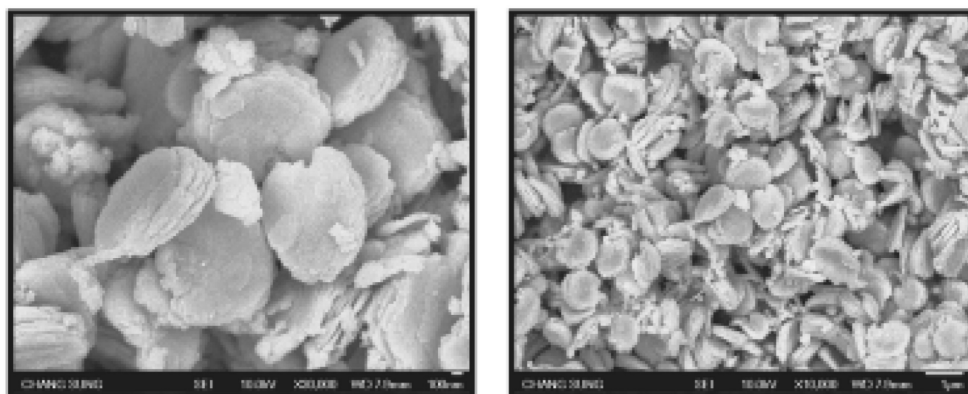


Fig. 10 SEM images of the hybrid filler system taken at 20,000x (left) and 10,000x magnification

als in the trend of changes in these industries. In addition, it is regarded as an important keyword for one step evolution of industry change. Technology of the UV curable PSA should also be evaluated in terms of complex to fit these changes. UV curable PSA will become even more important in the future. And it should challenge of integrating various technologies.

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ゴム科学—その現代的アプローチ—

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朝倉書店 (2016)

最近, ソフトマテリアルという言葉が闊歩している。かつて私が神戸大学で担当した高分子コロイド研究室もその後あっさり(?)「ソフトマター研究室」に変わった。コロイドもゴムもソフトな材料として共通性がある。本書は, 「まえがき: ソフトマテリアルとしてのゴム」に始まり, 序章まで読むだけで著者らの意図が明確に伝わってくる。章立ては次の通り。

- 1 序論 ゴムの歴史とその現代的課題
- 2 ゴムの基礎科学
- 3 ゴム・エラストマーの材料科学
- 4 ゴム・エラストマー技術の新展開
- 5 ニューマチックタイヤ
- 6 ゴム・エラストマー科学の未来

タイヤについての第5章は単著であるが, 他の章は複数の著者の分担あるいは討論の成果であるようだ。このような執筆形態では表現にばらつきが出て読み難い場合があるが, 本書はそれを感じさせない。推敲に十分な時間を取られた成果であろう。著者の言を借りれば, 本書は21世紀におけるゴム技術の新展開に対応すべく「現代的な」ゴム科学のテキストとして執筆され, また, ゴムの科学と技術につ

いての全般的な解説ではなく, 現代的分析技術の導入, 反応解析・材料設計, 工学的な製品設計の最新の成果の紹介に重点を置き, 体系的な解説がなされている。

高分子科学の確立に先だってゴム科学の成立があり, さらにその前に自動車用ゴムタイヤの大量生産というゴム工業の興隆があった。つまり, 技術が科学を先導した典型的な例であり, 私が長年携わってきた乳化重合による高分子エマルション・ラテックスの研究と共通するところがある。それ故, 著者らの思いに共感出来るのは, 新しい研究・解析手法による新たな発見・解明がゴム材料をさらに魅力あるソフトマテリアルと感じさせてくれる点にある。各章に配置されているコラム欄も興味深く, 著者らの哲学が感じられる。さらに, 「あとがき」にはゴム科学をさらに学習するための道しるべが, 書名を挙げ解説(批判?)されていて, 新たにゴム科学に関わる研究者や技術開発に携わる技術者には大変参考になる。

長年にわたり, ゴム科学の研究に没頭されてこられた著者らの紺碧の玉稿である。是非, 一読をお勧めしたい。

(南京工業大学特聘教授・神戸大学名誉教授 大久保 政芳)