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A Review on Electrically Conductive Adhesives in Electronic Packaging

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Abstract: ***Electrically conductive adhesives (ICAs; Isotropic Conductive Adhesive, ACAs; An- isotropic Conductive Adhesive &NCAs; Non-conductive Adhesive) offer interconnection of materials which can be used in electronic packaging to replace the traditional solders due toits advantages such as eco-friendliness, simple applying process, low curing temperature, and cost-effectiveness. ICAs have been developed and widely used in replacing traditional soldering, especially in die attach, surface mounting technologies and flip chip assemblies. ACAs can utilise in emerging electronic technologies like, multilayer printed circuits, thin- film transistors, smart card application etc. An electrically conductive adhesive (ECA) consist of an adhesive matrix in which the filler materials are embedded. Adhesive matrix offers towards binding properties of the ECA, where the filler materials contribute towards electrical, mechanical and thermal properties of ECAs. Apart from variety of conductive fillers incorporated into the matrix, orientation and distribution of fillers within the adhesive matrix also plays a key role in determining the overall properties of the ECAs. Processing methods as well as properties of conductive fillers like, vapour grown carbon nanofibers, (VGCNF), modified forms of graphene (AgNPs/rGO and nitrogen doped graphene), carbon nanotubes, conductive polymers and conventionally used metallic fillers (copper, silver, copper-silver alloys) are reviewed.***
Keywords: Electrically conductive adhesive, Electronic packaging, Conductive fillers

I. INTRODUCTION

Electronic packaging is the technology of electrically and mechanically interconnecting of components into an integrated system. The electronic package provides the conductive path required to achieve connection from one circuit element to another, provides protection of devices from the environment and enables removal of heat from devices. The primary function of the electronic package is electrical interconnection of components for distribution of electrical signals and power. Traditionally the interconnection within an electronic component has been made using copper circuitry and typically circuit traces for connections within a wiring plane. Nowadays interconnection among components (assembly) is commonly made using solder, most often tin–lead alloys. Tin–lead solders have been used as interconnecting materials in electronic packaging for many years. Tin-lead alloys have good electrical and mechanical properties, low melting point and good reliability, but they are not eco-friendly. Tin–lead solders contain lead, which is a hazardous material to human and environment. As the quantity of waste related to electronic products increases day by day, global legislative initiatives and corporate marketing strategies are driving a reduction in the use of such toxic substances in electronics. One solution is the use of lead-free solders in electronics [1]. Lead-free solders and electrically conductive adhesives (ECAs) have been considered as the promising alternatives of tin-lead alloys in recent years. Electrically conductive adhesives are generally composite materials composed of insulating adhesive binder resin that provides physical and mechanical properties such as adhesion, mechanical strength, impact strength and conductive fillers(such as, silver, gold, nickel or copper) that conducts electricity [2]. They can offer numerous advantages, such as environmental friendliness, mild processing conditions, fewer processing steps that can eliminate fluxing and reduce cleaning of the components which lowers processing cost, enable use of heat- sensitive and low cost components and substrates possibly, and fine pitch interconnect capability [3].

ECAs can be synthesized by employing two methods; the first one is where conductive fillers are dispersed within an insulating polymer matrix, and the second one is where inherently conductive polymers are blended with an insulating polymer matrix. Generally, conductive fillers can include metal fillers, carbon fillers, ceramic materials and metal-coated fillers, etc and inherently conductive polymers cover polypyrroles, polyanilines, polyacetylenes, etc. which can be mixed with an insulating polymer matrix. ECAs are divided broadly into two types of adhesives such as isotropic conductive adhesives (ICAs) and anisotropic conductive adhesives (ACAs) based on their morphology, filler percentage and properties [4]. ICAs, offer electrical conductivity in all x-, y- and z-directions due to high filler content exceeding the percolation threshold. For ACA or NCA, the electrical conductivity is provided only in z-direction and not on the x-y plane [5]. ECAs based on epoxy resins have attained widespread

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usage in electronic applications such as die attach adhesives, flip chip interconnection, etc. owing to their high chemical and thermal resistance, excellent mechanical properties, good adhesion to various materials, low shrinkage, availability of solvent free formulations, light in colour, ease of controlling viscosity, low vapour pressure, etc [4]. This review paper consists of the fundamentals of electricallyconductive adhesives focusing on the various types of ECAs and different types of conductive fillers based on metals, ceramics, carbon, metal coated fillers and conducting polymers. Future prospects and application of electrically conductive adhesives are also reviewed.

II. ELECTRICALLY CONDUCTIVE ADHESIVES (ECAS)

Conductive adhesives, which represent an intrinsically clean, simple alternative in electronic applications can be defined as composite materials comprising of solid conductive fillers dispersed in a insulative polymer matrix. When the polymer matrix is cured, it provides the mechanical adhesion between the respective substrates. A critical concentration of conductive fillers(percolation threshold), has been frequently observed above which the electrical conductivity of composites increases abruptly. This is where the fillers form a conductive network by connecting to each other within the base material thus providing a path for electron transport. Conductive adhesives are a promising lead-free alternative to SnPb soldersin applications like integrated circuits [4]. ECA can be categorized with respect to conductivefiller loading level into anisotropically conductive adhesives (ACA, with a typical 3–5 mm sized conductive fillers, or sometimes in a filmform, called aniostropically conductive film (ACF)) and isotropically conductive adhesives (ICA, with 1–10 mm sized fillers). The difference between ACA and ICA is based on the percolation theory which depends on the quantity of conductive fillers used. The percolation threshold depends on the shape and sizeof the fillers, but typically in the order of 15–25% volume fraction [6].

In addition to ACA and ICA, recently non-conductive adhesives (NCAs) bonding technology has been introduced, which requires no fillers and instead a relatively high bonding pressure coupled with heat to enable bonding between the IC chip and the substrate. When applying the heat and pressure, the direct and physical contact between the two surfaces of the IC bump and the substrate bond pad can be made and a permanent joint is formed by NCA resin curing/solidification [6].

III. ISOTROPIC CONDUCTIVE ADHESIVES (ICAS)

ICAs are being used to replace the traditional tin-lead solder alloys in electronic packaging and interconnects. ICAs are composites made with polymer resin and conductive fillers. The polymer resins, thermoplastic or thermosetting resins, are generally cured at high temperatureand provides the shrinkages force, low thermo-mechanical fatigue, adhesion strength, good chemical and corrosion strength. Thermosetting resins such as epoxy, silicone, polyurethane and thermoplastics such as phenolic epoxy, polyimide are common used isotropic conductive adhesive matrix resin. Conductive fillers include silver (Ag), gold (Au), nickel (Ni), copper (Cu) and tin, SnBi or SnIn coated Cu in various sizes and shapes. Silver is the most common conductive fillers for ICA due to high conductivity and easy process, but for the wide use of Ag-filled ICA application, its high cost is one of the drawbacks [2]. The main advantages of ICAs over tin/lead solders include:

A. They provide an eco-friendly alternative to solders as they're lead-free.
B. Since no flux is utilized in ICAs, the surroundings polluting Chlorofluorocarbonsolvents used to smooth the flux are eliminated.
C. They contain low processing temperature compared to soldering, which involvesexcessive temperature.
D. They have a low thermo-mechanical fatigue.
E. They may be used on a wide variety of surfaces (such as ceramics, glass and other non-solderable surfaces).
F. They may be applied for finer pitch packages like display screen printing of PCBs etc.
G. ICAs have the advantage for shorter and faster assembly times, because it involves fewer processing steps.
H. ICAs are basically low cost because the quantity of material used is substantiallyreduced [5].

A. ICAs for Surface Mount Technologies

Surface-mount technology (SMT) is the primary approach for interconnecting chip components to substrate. This process is carried out with the aid of using packing and setting the additives on the broadcast circuit board and the use of the reflow furnace to soften the solder alloy for the electronic system interconnections. As a potential replacement for lead- containing solders in SMT application, significant progress has been made to address different materials properties and reliability issues for the development of high performance ICAs. Due to some challenging issues ECAs cannot be used as drop-in replacements for solder in all applications, therefore tin–lead solder has been exclusively used as the interconnection material in surface-mount technology nowadays [7].

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B. Die (chip) Attach Adhesives

The primary function of die attach adhesives is to mechanically and electrically attach the integrated chip (IC) to the substrates. A good die attach material must have attributes suited to the desired functionality such as mechanical, thermal, and electrical properties. The die remains fixed in place when it is subjected to assembly processing or during actual device service, so that sufficient adhesion is required. Eutectic die connect is a low throughput, manual technique that may not be easily adapted for excessive speed automation. Although silver/glass die attach has provided some improvements over the eutectic technique, it still requires a lengthy and unique temperature profile to be able to get rid of the organic vehicleat a controlled rate. Non-uniform die attach can enhance die stresses (inflicting the die cracks), thus turning into a reliability issue. Polymer adhesives are used considerably with the attachment of IC to a variety of electronic packages. The polymer-based adhesives provide many benefits including lower stresses on IC die because of low material moduli and curing temperature, ease of use in a production environment, and low price in comparison toinorganic adhesives [6].

C. ICAs for Flip Chip Interconnects

Isotropic conductive adhesive materials use much higher loading than ACAs to give electrical conduction in all directions throughout the material. To be used for flip chip applications, it isnecessary to apply them on the areas which are to be electrically interconnected, and to make ensure that spreading of the materials do not occur during curing which would purpose electrical shorts between the different pathways. ICAs are usually provided in paste form. To exactly deposit the ICA paste, screen or stencil printing is most usually used. However, to try this to the size and accuracy required for flip chip bonding could require very correct sample alignment. The transfer method may be used to overcome this requirement. Raised studs or pillars are required on either the die or the substrate for this technique. The ICA is then selectively transferred to the raised region by contacting the face of the die to a flat thin film of the ICA paste. This thin film can be produced by screen printing and the transfer thickness can be balanced by controlling the printed film thickness. This method confines the paste to the area of the contact surfaces of the die and the amount can be adequately controlled so asto prevent spreading between pathways when the die is placed. Pressure during bonding isnot necessary in this process [7].

D. Improvement of Electrical Conductivity of ICAs

Polymer–metal-based electrically conductive adhesives (ECA) typically have lower electrical conductivity than Sn/Pb solders. To enhance the electrical conductivity performance of ECAs, different methods have been conducted and significant improvement of conductivityof ECA has been achieved.

1) *Increase of Polymer Matrix Shrinkage:* ICA pastes exhibit non conductive property before cure, but the conductivity increases after cured. During the polymer curing process, the ICAs achieve its electrical conductivity because of the shrinkage of polymer binder. Accordingly, ICAs with high cure shrinkage normally exhibit better conductivity. With increasing crosslinking density of ECAs, the shrinkage of the polymer matrix have been increased dramatically and subsequently the decreased resistivity of ECAs was observed. Therefore, the electrical conductivity increases due to the cure shrinkage of a polymer binder. The electrical conductivity for epoxy-based ICAs, can be increased with the addition of a small amount of a multi-functional epoxy resin into an ICA formulation and hence increase cross-linking density and shrinkage, [6]-[7].
2) *In Situ Replacement of Lubricant on Silver Flakes:* An ICA is generally consists of a polymer binder and silver flakes. A thin layer of organic lubricant is applied on the silver flake surface. The added lubricant layer performs a vital role in the performance of ICAs, such as the dispersion of the silver flakes within the adhesives and the rheology of the adhesive formulations of the composite. This organic lubricant layer, is a typically fatty acid such as stearic acid that forms a silver salt complex between the Ag surface and the lubricant [6]. By in-situ replacement of the surfactant-stearic acid in silver (Ag) flakes, such difunctionalacids can increase the conductivity of ECAs [8]. A suitable lubricant remover is a short chain dicarboxylic acid since its strong affinity of carboxylic functional group (–COOH) with stronger acidity of such short chain dicarboxylic acids and silver. The conductivity of an ICA can be improved significantly with the addition of a small quantity of short chain dicarboxylic acid because of the easier electronic tunnelling with the aid of using the intimate flake–flake contacts in the silver flake networks [7].
3) *Incorporation of Reducing Agents:* Silver flakes are the most used fillers for conductive adhesives due to the high conductivity ofsilver oxide as compared to other metal oxides, most of which are non-conductive. However, the conductivity of silver oxide is not as good as the metal itself. The electrical conductivity of ICAs will be improved with the use of reducing agents. Aldehydes have been added into a typical ICA formulation and improved conductivity was achieved due to reaction between

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silver oxide and aldehyde that exists on the surface of metal fillers in ECAs during the curing process:

$$R–CHO + Ag2O \rightarrow R–COOH + 2Ag. \quad (1)$$

The oxidation product of carboxylic acids, aldehydes, which are stronger acids and have shorter molecular length than stearic acid, may also partially replace or remove the stearic acid on silver flakes and contribute to the improved electrical conductivity [7].

4) *Low-Temperature Transient Liquid Phase Fillers:* For improving electrical conductivity the another method is to incorporate transient liquid- phase metallic fillers in ICA formulations. The filler used in this formulation is a mixture of a high-melting-point metal powder (such as Cu) and a low-melting-point alloy powder (such as Sn–Pb or Sn–In).During the cure of the polymer matrix, the low-melting-alloy filler melts when its melting point is reached. The high melting point particle will dissolve in liquid phase. The liquid exists only for a limited period of time and then forms an alloy and solidifies. The electrical conduction is maintained through a metallurgical connections in situ formed from the metal and alloy powders in the polymer binder [6].

E. Reliability Enhancements of ICA Interconnects

Critical reliability issues of ICA joints in electronic packaging applications are mainly because of the unstable contact resistance between ICA and metal finished components under environmental attacks, includes humidity and temperature cycling/aging. For excessive temperature and humidity growing environment, the galvanic corrosion rather than simple thermal oxidation at the contact area between metallic fillers in ICA and non-noble metal finish is known as the most underlying mechanism for unstable contact resistance. Therefore, most research works for improving the stability of electrical conductivity of ICA joints have focused to minimize the unstable contact resistance mechanism of ICA joints [7]. Several possible methods are: uses of oxygen scavenger, corrosion inhibitors, development of polymer resin with low moisture absorption, Ag coated Cu flakes [9], in-situ replacement of the surfactant-stearic acid in silver(Ag) flakes [8], also for the reliability improvement of Silver(Ag)-based ICA joints, migration of silver (Ag) is most serious concern.

IV. ANISOTROPIC CONDUCTIVE ADHESIVES (ACAS)

Environmentally-friendly manufacturing is an important goal for the electronic packaging industry in the modern era. Using anisotropic conductive adhesives (ACA) instead of soldering helps achieve these goals. The use of ACA for direct interconnection of flipped silicon chips to flip chip packaging (aka printed circuit) offers advantages such as lower assembly process temperature, low cost, smaller thickness. It is a composite composed of fineconductive particles that are uniformly dispersed in a thermosetting epoxy matrix and have been widely used for interconnecting microelectronic elements. There are mainly two forms: (1) paste (2) an anisotropically conductive film (ACF) [10]. The paste form adhesives are been sandwiched between the substrate to bond them together supplied by heat and pressure [4]. ACFs are produced with the help of a random distribution of conductive fillers enough to provide electrical conductivity in Z-direction and few fillers are available to restrict the conductivity in X–Y direction [6].The ACF assembly process consists of an adhesive resin and finely conductive fillers such as metal particles or metal-coated polymer [13] and requires the application of pressure while the adhesive cures [6].

It has been widely used in flat panel display modules to provide better resolution, lightweight, narrow profile, and lower power consumption in the film forms, and their electrical conductivity is provided only in z-direction between the electrodes, where chip surface that uses thermally conductive ACA heats up faster than traditional ACA seals, which means that the thermally conductive adhesive dissipates heat from the source more easily than traditionalthermoplastic adhesive [2]. ACAs have application that have flat panel display (ACF), smart card, flip-chips board and flip-chip on glass, where solder can't be used due to heat sensitivity [4].

A. Materials and Processing

They can be classified as unidirectional ACAs or bidirectional ACAs based on their conductivity path, which can be achieved by applying external forces and being supplied as either thermosetting or thermoplastic materials liquids and pastes which can be cured by heat or used as single component adhesives [4]. Two basic types of adhesives are available: thermosetting and thermoplastic materials. Thermosetting adhesives such as epoxies and silicones form a three-dimensional cross-linked structure when cured under certain conditions equipment, the curing techniques includes UV light, catalyst, heat. Due to this initial uncross-linked material is modified into a rigid solid and this process is not reversible. Thermoplastic adhesives are rigid materials, below theglass transition temperature of a polymer. The principal advantage of thermoplastic adhesivesis the relative ease with which the interconnection can be disassembled for repair operations. Above the glass transition temperature, they exhibit flow characteristics and to achieve good adhesion, the assembly temperatures must exceed the glass transition temperature.

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During theprocess, pressure is been provided during the adhesive curing and adhesive need to besupplied by energy to initiate the chemical reaction, mostly heat and UV radiation (through optical fibre bundle) are used for this purpose.
Important process parameters for ACA/ACF assembly are pressure load, temperature, tackingtime (time required for the adhesive to soften and flow), and bonding time. During the process the interconnecting parts are preheated below the bonding temperature, hence the films soften up and able to fill void areas. The bonding load must be high enough for the conductive balls to make fine physical contact between the conductors, but not high enough to damage any of the parts. The bonding quality depends upon certain parameter such as bonding temperature, curing temperature, alignment accuracy, pressure application and value [6].

B. ACA/ACF Applications and Limitations

ACA/ACF has applications in flex circuits and surface mount technology (SMT) for the chip-scale package (CSP), application-specific integrated circuit (ASIC), and flip-chip attachment for cell phones, radios, personal digital assistants (PDAs), and memory chip in laptop computers. Some key issues hinder their implementations as high-power devices. The ACA/ACF joints generally have a poor current carrying capability, lower electrical conductivity, and electrical failure during thermal cycling.
The LCD are been assembled by tape carrier package (TCP) interconnection technologies in which it is divided into flat type and bending type. The flat type makes LCD thinner, but hasa difficulty to reduce the frame size, and this limitation is eliminated by bending type, which makes the module smaller. For better thermal bonding, a mismatch is provided between the TCP and the panel. TCP extends more than the panel after the adhesive of the ACF is cured and extension is retained. Therefore, the correction value should be considered conduct the alignment. Chip on flex (COF) uses tape-automated bonding (TAB) of the ICs for connecting the liquid crystal panel and the printed wiring- boards (PWBs) in a bridge form are popular for packaging of LCDs. The chip on flex (COF) technologies using ACF would overcome the limitation of the fine pitch capability in tape carrier package (TCP) by making use of thin Cu (copper) foils and low CTE base film materials. Therefore, the outer lead interconnection of COF using ACF has been of much interest because it provides a fine pitch capability smaller than 50 mm pitch. With a chip on glass (COG) packaging, bare chips are connected directlyto the liquid crystal panel, this packaging can minimize connection pitch by flip-chip, face- down, IC bonding. However, it has a disadvantage that the mounting of ICs on the LCDpanel requires additional area for the IC chips and bus lines, thereby expands the peripheral area of the display to reduce the space allocated for packaging by concentrating the positionof the driver ICs and by changing its length-to-width ratio [6].

C. Improvement of Electrical Properties of ACA/ACF

ACA joints are determined directly by mechanical contact between chip connections and electrodes on chip carriers. The bonding force plays a crucial role in electrical performance. Therefore, high bonding pressure is certainly favourable for close contact and thus for loss in Contact resistance. The improvement of the organic self-assembly monolayer and the low- temperature sintering of ACA filled with nano-Ag are two methods for improving the electrical properties [6].

1) *Organic Self-Assembly Monolayer Enhancement:* Organic monolayers have been introduced at the interface between metal filler and metal- finished bond pad of ACAs, in order to enhance the electrical performance of ACA material. The electrical resistance is reduced and provides high current flow due to the organic molecules, which bonds to metal surface and form physic-chemical bond. The unique electrical properties are because of their tuning of metal work functions by these organic monolayers [7]. The mainly used organic monolayers are dicarboxylic acid and dithol, introduced into the ACA joints for gold-filled ACAs and silver-filled ACAs. The low temperature curable ACA (<100°C) is achieved by dithol incorporated ACA with micron- sized gold fillers, which lower the joint resistance and elevated maximum allowable current. The high curing temperature ACA (150°C) is achieved by dicarboxylic acid was introduced into the linkage of nano-silver filled ACAs, which significantly improves electrical properties[6].
2) *Low Temperature Sintering of Nano- Silver(Ag) Filled ACA:* Melting temperature (Tm) of Ag and sintering temperatures of materials could be reduced by lowering the size of the materials hence for this sintering behaviour must occur at lower temperature, at which the fine metal particle ensures high electrical performance by eliminating the bonding between metal fillers. The nano sized particle increases the number of conductive fillers; results in more contact surface are between fillers and bond pads. The Current–resistance (I–R) relationship of the nano-Ag filled ACA shows that the resistance of ACA joints decreased dramatically, with increasing curing temperature. Also, ACA samples exhibited higher current carrying capability at higher curing temperature than the low temperature. This suggests that increased sintering of nano-Ag particles results in supercilious interfacial properties between fillers and metal bond pads were attained at higher temperatures, yet the x–y direction of the ACF has an outstanding dielectric property for electrical insulation [6].

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D. Improvement of Thermal Properties of ACA/ACF

For ACA joints to deliver high current, not only a low electrical resistance, high thermal conductivity of the interconnect materials is need to be required. In which it can helpdissipate heat more effectively from adhesive joints generated at high current. Therefore, a higher thermal conductivity needs to be achieved for an improved current carrying capability.

With the introduction of suitable organic monolayer treatment, improved interface property between metallic fillers and polymer resins could be achieved, whereby the thermal conductivity of ACA compounds was improved. It is also reported that by adding fillers with high thermal conductivity (carbon nanotubes) to the ACA formulation, the higher thermal conductivity could be achieved, which also provided a high transport capacity of additional. Carbon nanotubes, with high thermal conductivity (> 3000 W / K from theoretical calculations) can also be aligned within the composite matrix to improve the thermal conductivity [6].

V. CONDUCTIVE FILLERS IN ECAS

Generally, ECAs consist of matrix which causes the adhesive as well as binding properties of the compound, whose major drawback is that it's lack of electric conductivity. So as to impartelectrical conductivity, conductive fillers based on metals, ceramics, carbon, metal coated fillers and conducting polymers are embedded into matrix. These fillers are also observed to influence the mechanical as well as thermal properties of ECAs. Fillers used in recentresearch works and their effects on electrical conductivity are discussed below:

A. Vapour Grown Carbon Nanofibers (VGCNF)

A lot of studies have been undergone in recent years regarding the improvement of electrical mechanical and thermal properties of polymers containing Vapour grown carbon nanofibers as filler material (VCNFs). They have high aspect ratio and are available with diameters ranging from 50 to 200 nm and lengths of 50-100 nm.

Lower cost as well as close resemblance to conventionally used Carbon nanotubes (CNT) makes them more desirable even if their properties are inferior to CNTs. Hydrocarbons or carbon monoxide is subjected to catalytic vapour deposition over a metal catalyst for the production of VGNFs. Structure of VGNFs are described as tubular in nature consist of single or double stacks of truncated graphite cones which intersect fiber axis at very highly oblique angles along central hollow. Depending upon the cone angle and stacking, various structures of the fibres are possible including bamboo like and cup-stacked. Vapour deposited carbon are distributed over the outer surface of carbon fiber which causes the bonding as wellas the thickening of fiber in larger aggregates. The exterior and interior layer of fiber varies in graphite concentration and observed to be consists of disoriented embryonic graphene planes.

The graphitised VGCNF exhibits very high electrical and thermal conductivity values for reinforcement in polymers. The VGCNF/epoxy composites were prepared by three different mixing techniques.

1) Conventional mechanical mixing (MM)

2) Combined sonication and mechanical mixing (CSM)3.Three-roll mill mixing (RM)

The effect of these processing methods on the electrical conductivity is observed as the highest increase in electrical conductivity with VGCNF loading is observed for the composites produced by the RM and the compound produced by CSM has observed the lowest conductivity among the three. The percolation threshold (Pc), defined as the filler content required to achieve a conductivity of P >10 ^-6 Sm^-1, for VGCNF/RE composites produced by RM was found to be just less than 2 wt.%. On the other hand, composite produced by MM at 4 wt % has high value for Pc, where else composite produced at 2 wt% are non-detectable by the device .For 12 wt.% VGCNF/RE composite produced by MM, no significant increase in electrical conductivity is observed due to increase in mixing time at 4500 rpm in contrast to the thermal property of the composite produced by MM, which is because a speed of 4500 rpm (for no more than 15 mins) was used for all the composite produced by MM. However, it should be noted that unlike thermal conductivity, the electricalconductivity of 12 wt.% VGCNF/RE composite produced by RM is an order of magnitude higher than that of the corresponding composite produced by MM. Increase in electrical conductivity as well as the lower pc obtained for the composites in RM indicates the uniform dispersion as well as the distribution of VGCNF in the matrix, which results in increase of electrical conductivity of resulting composite due to probability in increased electron tunnelling as well as decrease in separation by polymer matrix. The 10 wt % VGCNF composite produced by MM is one order of magnitude greater than the corresponding composite produced by CSM. Either breaking of carbon bridges between the fibers or decrease in aspect, occurs during sonication can be the reason behind. No increase in electrical conductivity is observed due to increase in sonication time for 10 wt % of VGNCF/RE compounds produced by CSM method and this is similar to what is observed for its thermal conductivity. The electrical conductivities of VGCNF/RE composites produced by RM are almost the same in every direction at all loadings of VGCNFs.

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This clearly indicates that the composites produced by RM have no preferred orientation of the VGCNFs, which is thus oriented randomly in the matrix. Hence out of all composite produced, the composite produced by RM are isotropic. Where else the electrical conductivity in z direction is slightly lower than that of the electricalconductivity along x or y direction for composites produced by MM method, which point towards the slightly better orientation of fibers up to 15 wt % loading of VGCNFs in x or y direction. These results further indicate the higher isotropic nature of composites produced byRM rather than the composite produced by MM [14].

B. Ag-Coated Cu Flakes

Isotropically conductive adhesives (ICAs) have been identified as one of the promisingalternatives for eutectic Sn/Pb solders in electronic packaging as they offer many advantages over conventional solders. These advantages include, low stress on the substrate, low processing temperature, environmental friendliness and flexible and simple processing. ICAs are composites of conductive fillers and polymer matrices, where the polymer matrices, such as epoxy, provide adhesive bonding between a component and a substrate where electrically conductive fillers provide electrical conduction. Among various electrically conductive fillers, silver is the filler of choice in the industry because of easy processing and excellent electrical and thermal conductivities. However, one of the main impediments in the wide application of silver into silver filled ICAs in microelectronics are its high cost. For low cost electrically conductive fillers copper is the most promising candidate, but dramatically increase in the resistivity of copper-filled ICAs due to corrosion and oxidation considerably limits their applications. To enhance oxidation resistance of copper particles, the coating of copper particles with Ag has been investigated .It is reported that a Ag coating on copper flakes provided good oxidation resistance at temperatures lower than 175°C; however the resistivity of ICAs filled with the Ag-coated Cu flakes increased from 1.6×10^-3 ohm cm. Before the heat treatment of the fillers to 2.5 ×10^-3 ohm cm after the heat treatment at175°C for 2h as a result of the formation of Cu2O on the Ag-coated Cu flakes [26]. The oxidation of Ag-coated Cu flakes may result from incomplete plating of the copper flakeswith Ag particles, investigated the effect of silane coupling agents and dispersing agents on the corrosion stability and electrical conductivity of silicone sealants filled with Ag-coated Cu powders. With the most effective silane coupling agent, 3-aminopropyl-Triethoxysilane, silicone sealants filled with the Ag-coated Cu flakes showed a 30% increase in the resistivity after storage at 50°C for 24 h. These results indicate oxidation of the Ag-coated Cu powdersat high temperatures. Additionally, when the moisture and oxygen come in contact with the exposed Copper and silver interface at the edge of the pits, galvanic corrosion can occur in pits in the silver plating resulting in difference in electrochemical potential between. Therefore, the prevention of oxidation and corrosion of exposed copper in Ag-coated Cu flakes is crucial for the preparation of highly reliable ICAs filled with Ag-coated Cu flakes [15].

To retard or prevent the oxidation and corrosion of copper, various organic corrosion inhibitors have been widely used. These inhibitors include azoles, amines, amino acids and other compounds. They form a complex with copper that acts as a barrier film between metal and environment, upon absorbing on to the metal surface. However, the problem is that these excellent corrosion inhibitors are thermally unstable at elevated temperatures, and excess corrosion inhibitors can degrade the electrical and mechanical properties of ICAs [9].

One of the main hurdles for the wide use of current Ag-filled, isotropically conductive adhesives (ICAs) is the high cost of silver fillers, while the challenge for low cost copper- filled ICAs is their poor reliability. Here low cost, highly reliable ICAs in which the copper flakes used as filler are coated with Silver (Ag-coated Cu flakes) have been developed. With silver-coated copper flakes modified by an amine-based silane coupling agent (SCA), the ICAs with the resistivity comparable to that of commercially available silver filled ICAs havebeen achieved. Moreover, the contact resistance of the ICAs filled with the modified Ag- coated Cu flakes on a Ni/Au surface can be stabilized for more than 1000 h of aging at 85°C and 85% RH and after three reflows with a peak reflow temperature of 255°C. The causes leading to the better reliability and higher conductivity of the ICAs filled with Ag-coated Cu flakes modified by amine-based silane coupling agent are observed [9]. By adding suitable trace elements such as Ag and Mg to Cu fillers, Cu can be promising candidate conductive metallic filler for ECA [8].

C. AgNPs/reduced Graphene Oxide (rGO) Nanocomposites

Graphene is a quasi-two-dimensional (2D) structure material, in which monolayers of carbon atoms are packed into honeycomb crystal planes. Graphene is a material having its own unique mechanical, thermal as well as electrical properties. However, the graphene sheets were also aggregated due to its vanderwaals force and strong π-π interaction. To explore the potential of graphene, many efforts have been made to functionalize graphene with metal nanoparticles. A large number of metal nanoparticles were assembled on the surface of graphene oxide sheets based on large specific surface area.

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And nano-sized silver particles can prevent graphene sheets from aggregating, so the hybrids nanocomposites are particularly attractive in terms of their distinctive optical and electrochemical properties, high catalytic activity, and strong surface-enhanced effect. With the development of electronics packaging technology, chip packaging size is getting smaller and smaller, and the unit power in chip consumption has increased dramatically however, most of the power changes into heat, whichaffects the longevity of electronic devices as well as reliability of chips due to the rising chip temperature. Therefore, the thermal management of the electronics packaging is also of great importance [16]. AgNPs with the size from 15 to 20 nm and narrow size distribution, which were successfully decorated on rGO sheets by a one-pot method in aqueous solution. Then ECAs were prepared by mixing AgNPs/rGO, silver flake and epoxy. The optimum content of conductive filler and electrical properties of ECAs were studied after cured at 150°C.

Electrically conductive fillers are dispersed into the matrix resin to form electrically conductive channels and achieve electrically conductive connects. But fillers can't contact with each other completely after only doped micron silver flakes in the conductive adhesive because micron silver flakes formed many gaps and voids. It decreased the number ofelectrically conductive channels. AgNPs/rGO nanocomposites can fill into these gaps and voids and thus formed new channels and exhibit excellent electrical conductivity. Relationship between AgNPs/rGO content and electrical resistivity with 70 wt% of the total content, the electrical resistivity increased after the first decreased with the increase of AgNPs/rGO mass fraction. The electrical resistivity fell to 8.76 ×10^5 ohm cm when the mass fraction of AgNPs/rGO reached to 0.2 wt%. When the mass fraction exceeds 0.2 wt % electric resistivity is observed as increasing. It was attributed to uneven dispersion and agglomerated of AgNPs/rGO nanocomposites filler in the ECAs at higher loading [17].

D. Nitrogen doped Graphene Nanosheets

Nitrogen-doped graphene nanosheets (N-GNSs) were used as a performance improver for a silver-filled electrically conductive adhesive (ECA) as well as a conductive filler for apolymer resin adhesive. The N-GNS samples were prepared by the chemical-intercalation/thermal-exfoliation of graphite followed by a thermal treatment in NH3. Only 1 wt.% of N-GNSs was required for the adhesive to reach a percolation threshold, and the performance using N-GNSs was much better than that obtained using carbon black or multi- walled carbon nanotubes (MWCNTs). An investigation on the effect of N-GNS or MWCNT over silver flakes filled ECAs upon lower silver loading regarding reduced electrical resistivity was studied. With 30 wt.% of Ag filler, the polymer resin was still non- conducting, while a resistivity of 2.2×10^-3 ohm cm was obtained using an Ag/N-GNS hybrid filler fortified with only 1 wt.% of N-GNSs due to large specific surface area, high aspect ratio and good electrical conductivity of the doped graphene.

Upon comparing the electrical resistivity of different GNS representing the ECAs filled with N-GNSs prepared in different ammonia flow rates, the resistivity clearly decreased with the increase in the NH3 flow rate used. And the study revealed that nitrogen doping significantly improved the conductivity of graphene and reduced the resistivity of graphene-filled ECAs by up to two orders of magnitude. The nitrogen content on graphene, and hence the conductivity of the ECAs, increased with increasing NH3 gas flow rate [18].

E. Carbon Nanotubes

Carbon nanotubes (CNTs) are the most anticipated next generation highly conductive adhesive electrical wires having additional benefits of low cost, high thermal conductivity, low weight, as well as high mechanical performance. Due to the versatility in electrical and physical properties, CNTs causes intrinsic electrical resistivity and are able to lower percolation thresholds in matrix. Moreover, CNTs pocess very high aspect ratio, which would be applicable for connecting metal flakes in ECAs for constructing electrically conductive networks and reducing the cost of ECA by the reduction of metal content. Therefore, CNTs could be one of the most promising fillers for low cost ECAs [19].

Ag flakes and CNTs were used as co-fillers to build the conductive network because CNTs asconductive nanomaterials could bridge the neighbouring Ag flakes to accelerate the electron transport [19]. Lower electrical resistance as well as higher mechanical strength where indicated by the results as compared to the conventional fillers [20]. When the filler content exceeds a critical volume fraction, formation of conductive pathways results in the occurrence of electrical conductivity. At low filler contents the, ability to conglomerate, its dispersibility and aspect ratio of filler are crucial parameters in the occurrence of electrical conductivity . Consequently, due to the relatively low surface area and high aspect ratio, MWCNTs exhibit the highest potential for an efficient enhancement of the electrical conductivity. Any kind of treatment, leading to a reduction of the aspect ratio (functionalization, ultrasonication, etc.) correspondingly increases the percolation threshold [21].

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F. Copper Flakes

Copper is considered as promising filler material for ECAs due to its improved electro- migration performance, low resistivity as well as its lower cost consideration. Depletion of electrical conductivity is observed in copper due to the oxidation of copper at lower temperature, which arises due to the lack of self-protective layer to prevent further oxidation. Electrical resistivity as well as thermal stability of ECAs has been studied recently on the basis of copper fillers with different morphology and particle size. Wet chemical reduction, electrolytic and gas atomization method, respectively are the methods implemented for the preparation of copper filler used in this experiment. Variation in electrical resistivity is observed for cured ECAs filled with different type of copper fillers. Enhanced formation of conductive channels and increased conductive networks in ECAs is observed to be leading to reduced electrical resistivity, which can be achieved by the cooper filler with smaller particle size having higher packing density and larger surface area. It is also observed that low viscosity of ECA paste reduced electrical resistivity and enhanced shear strength of ECAjoint due to better surface wetting of Cu filler in the polymer matrices and enhanced surface wetting of the ECA paste to the Cu specimens of shear test [22].

Out of 3 different combinations of ECAs used, ECA Cu-3 shows the highest electrical resistivity among the other cured ECAs, whereas, electrical resistivity of ECA Cu-2 is comparable to that of the ECA Cu-1. The resistivity of the cured ECA Cu-3 was about 8 times higher than that of the ECA Cu-1 and times higher than that of the ECA Cu-2. The formation of conductive channels among the ECA fillers are the possible reasons for these effects observed. This may be due to the arrangement copper fillers inside the polymer matrix after curing. The packing density, particle shape, fractional size and spatial distribution of particles inside the polymer matrix etc. are the factors observed to be influencing the electrical conductivity of ECAs.

With same mass% (i.e. 80 mass%) of Cu fillers prepared in each ECA in this study, larger amount of fillers would be found side the ECA Cu-1 and ECA Cu-2 compared to that of the ECA Cu-3 due to relatively large particle size of Cu-3.Gap between adjacent particles can be reduced subsequently by reducing the size of dispersed particles. Thus, a relatively high packing density would be found in ECA Cu-1 and ECA Cu-2 compared with that of the ECA Cu-3.In addition, the morphology of the filler plays a key role in the ECA resistivity. So as to achieve low percolation threshold, fillers having elongated geometry like fibers or sheets can be used, due to the fact that fibers or sheets with higher aspect ratios (width-to-thickness) have great advantage over spherical or elliptical fillers in forming conductive networks in polymer matrix [23]. Cu is a promising candidate for conductive metallic filler owing to its electro migration performance, and low cost. In summary, we can conclude that the Cu fillers with different morphology have significant effect on the electrical resistivity in ECA [24].

G. Conductive Polymers

Polymer-based packaging materials are consist of polymer and filler, which combine the advantages of inorganic filler such as good thermal and electrical conductivity and the advantages of organic polymer such as easy processing and light weight. Most of the polymers that are used in polymer based packaging materials are thermosetting, such as epoxy, phenolic, polyester and Silicone. Polymer and its composite are the enabling technology, which can be used as adhesives to glue the semiconductor chips to a metal lead frame, encapsulation of chip by the mould compound after being mounded, the thermal interface materials (TIM) to dissipate heat, the embedded capacitive materials to storage energy and the electromagnetic interference shielding materials to attenuate the unwanted electromagnetic waves. However, polymer-based packaging materials generally have much lower electrical and thermal conductivities than those with ceramic-based and metal-based packaging materials. Therefore, more and more attention in the academic and industrial field has attracted towards the enhancement of electrical and thermal properties. For example, filler size distribution, modification of the resin matrix, optimization of fillers, optimization of fillers types, and the interface tailoring between the filler and the polymer matrix, those technology provide an effective way to improve the performance of the polymer-basedpacking materials [11].

The dielectric constants of polymers are strongly influenced by their chemical structure. Generally, molecules with more polar groups have higher dielectric constants. Polymers with very low dielectric constants (<3) such as polyethylene and polytetrafluoro – ethylene, do not always meet the combined thermal and mechanical requirements for application in electronic devices, nor do they have desirable adhesive characteristics. Polyimides with much better thermal and mechanical properties have dielectric constants between 3 and 4. Efforts to combine the desirable features of the Fluoro polymers with those of the polyimides have beensuccessful in that the resulting compositions have dielectric constants equal to or smaller than3 without sacrificing too much of the thermal and mechanical properties of the polyimides [25]. It is also observed that the electrical conductivity of the composite may be increased by the presence of electrically conductive fillers. However, in order to increase the thermal conductivity of the composite, the interfacial resistance between the polymer and the filler must be reduced [26].

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The resistivity of a conductive polymer composite is determined by thecomposite composition (such as filler loading), the surface properties of conductive fillers (such as the presence of a thin layer of lubricant or oxide film), physiochemical properties of polymer matrix (such as cure shrinkage and the interaction between the polymer matrix and conductive fillers), interlayer thickness, temperature, processing conditions of conductive polymer composites, etc. Reducing or even eliminating the contact resistance between conductive fillers is crucial for the preparation of highly conductive polymer composites [27].

VI.NON CONDUCTIVE ADHESIVES (NCAS)

The non-conductive assembly of components to the substrates is not often used method, but some experiments with these connections of components to substrates by NCA were already made as to the replacement of lead soldering, At present, the flexible substrates need flexible and low-temperature joints and for this applications, the soldering is not possible and NCA could be good was as an alternative to the ECA and this is the reason to realize thisexperiment [28]. Non-conductive adhesives, typically used in applications with smooth, well-defined surfaces such as flip chips, are available in paste or film form and are generally filled with silica or alumina to provide optimal flow properties for delivering and improvingthermal conductivities. The electrical contact is established through physical-mechanical contact [29]. Traditionally; the ACF only has one layer of the adhesive film along with conductive particles. Double layer ACF adhesive which has an additional non-conductive adhesive layer was introduced to reduce the electrical short failures in ultra-fine pitch applications the non-conductive adhesive layer composed of low viscosity thermosetting resin [30].

Hot melt adhesives are usually used for NCA bonding due to their easy handling. One commonly used adhesive includes the class of thermoplastic polyurethanes. In addition, conductive sheets can be used when trying to connect larger surfaces in shorter times. Electrical contact, pressure are exerted on the connectors during the joining process. The mechanical strength of these compression bonded parts is acceptable for most applications. NCAs have been used to bond chipped chips onto rigid or flexible substrates. Here the electrical connection is established by two conductive surfaces in contact without bonding material and the NCA only generates the permanent stress that keeps the surfaces in contact [29]. The mechanical shear strength for epoxy NCA and better mechanical shear strength for more adhesive amount.

On the other hand, the epoxy NCA and more adhesive amount have worse electrical resistance [28]. To fulfill low-temperature bonding, a commercial NCA that can be cured below 100degree Celsius was utilized. To minimize the thermal impact on the LCD module and flexible substrate, NCAs with a low cure temperature and reasonable cure time is preferred. During the thermal cycle, a high temperature would offer the possibility of additional curing of the NCAs, shrinking more and compressing bumps and pads, hence the decrease of contact resistance.

The NCA chip assembly is highly dependent on the performance of the NCA materials. The relaxation of NCA leads to the weakening of the compressive force provided by NCA, which would cause interfacial degradation and even destruction between bumps and pads [31].

A. Properties of NCAs

NCA joints avoid short-circuiting and aren't restricted, in terms of particle size or percolation phenomena, to a discount of instrumentality pitches. Any benefits embody cost-effectiveness, the easy process relating to the likelihood of non-structured adhesive application. For NCA the electrical conduction is additionally achieved through physical contact (although shaped underneath high pressure), and no metallurgic joints were shaped. Therefore, it's the unstable contact resistance drawback failures occurring in NCA joints that are caused by moisture- induced absorbent swelling and stress relaxation. Factors that have an effect on the degradation of the compressive force that maintains the mechanical contact within the NCA flip–chip structure content of the non-conducting filler may be a key issue that controls the fundamental properties of NCA materials. [6].

B. Future advances of NCAs

Adhesive that merges the electrical conduction along the z-direction (ACA) and the fine pitch capability (NCA-like) is emerging. The physical contact between the rough surfaces induces the electrical to interconnect and therefore the electrons suffer it. To confirm the standard NCA bonding, a comparative air mass (30 MPa) is required to deform the surface. The nano- sized semiconducting particles between the surfaces will improve conductivity and lower the applied pressure compared to a typical NCA method, leading to the sweetening of current- carrying capability [6].

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VII. APPLICATIONS

A. Compared with standard tin–lead solders, ECAs are more environmentally friendly, and that they are widely applied to electronic packaging and alternative industrial technologies, like liquid crystal display (LCD) and solar cells [18][32].

B. Silver-filled ICAs are widely utilized in a surface mount technology (SMT), flip chip, chip scale package (CSP), and ball grid array (BGA) applications. so many research efforts have been focused on the design and development of bio adhesives. These biomaterials will be utilized in various biomedical fields, including drug delivery, tissue engineering, wound hilling, and biosensors [33].

C. Poly(pyrrole-co-dopamine) [P(Py-co-DA)] or poly(aniline-co-dopamine) [P(ANI-co- DA)] or their composites attracted an excellent deal of interest because of their numerous applications in both industrial similarly as biomedical fields. It is worth noting that the copolymerization of Py or ANI with DA led to some biological features that are not available when homo-polymer was used alone. In detail, the copolymerization of DA with Py or ANI can improve solubility, biocompatibility, and adhesive properties [33].

D. ECA is believed to be more environmentally friendly with lower processing temperature, finer pitch capability, easier process, and greater flexibility compared with standard soldering-based technology [34].

E. ECAs can achieve low cure temperature, high line resolutions, environment friendly and meet the development on the miniaturization of electronic products and high integration of chip interconnection techniques on printed circuit boards or substrate [35].

F. In graphene nanosheet based ECAs because of high electrical conductivity make it applied in several areas like conductive membrane, catalytic, solar cells and super capacitor [35].

G. Today's blooming electronic technologies, like multilayer printed circuits, thin-film transistors (TFTs), transparent conductive coating, and solar cells require advanced materials that supply high electrical conductivity, good flexibility, proper stretchability, and satisfied mechanical strength. In addition, these materials should be of low cost, not harmful toenvironment, and easy process able. ECAs are considered together of the foremost feasible alternative interconnection materials for future applications ECAs [36].

H. In the previous couple of years, anisotropically conductive adhesives (ACAs) have gained increased popularity in flip chip packaging applications. Because of their anisotropic conductivity, these adhesives can be deposited over the entire surface, thus greatly facilitatingalso in material application [37].

I. In the development of thermosetting setting solder conductive adhesives by using Sn/Bi solder powder and polymer matrix [38].

J. Utilization of ECAs in electrochromic devices (ECDs) are widely utilized in smart windows, smart mirrors, goggles, displays, and auto-dimming rear-view mirrors. They have also attracted much attention as an emerging technology for next-generation applications like military camouflage and heat control of space satellites. For high-performance ECDs, the utilization of transparent conducting electrodes (TCEs) is important. Glass coated with indium tin oxide (ITO) has been widely used as an electrode component in ECDs given its high transparency and low sheet resistance [39].

K. Silver filled ECAs are widely used in a surface mount technology (SMT), ball grid array(BGA) applications, chip scale packaging(CSP), and flip chip [33].

L. ECAs are suitable to be used on polyester, poly-carbonate substrate, typical for use in smart card application [40].

VIII. FUTURE CHALLENGES

A. Repair is always important for a high volume manufacturing process. Knowledge needs to be generated about repair methods, repair reliability, and adhesive selection. Two key questions concern how to remove residues on the board when using thermosetting type conductive adhesives, and what are the reliability and processability if a new adhesive dot is applied on top of the residue.

B. Poor adhesion to the metal surface of conductive adhesives is one of the most intractable problems when they are used for high volume surface mount applications.

C. At present, knowledge is lacking about how to correlate accelerated testing data with actual lifetime because of the composite nature of the conductive adhesive joint. Consequently, models available for soldered joints and for pure polymers cannot be applied directly to conductive adhesive joints.

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Despite these challenges, conductive adhesive joining is believed to be most promising for directly bonding barechips on both flexible and rigid printed circuitry. Compared with soldering, this joining approach results in a cost effective and reliable system. A total electrical connection system using a single metal is achievable with conductive adhesives, thereby avoiding problems caused by the formation of intermetallics when more than one metal is present [12].

IX. CONCLUSION

This review article have discussed about various aspects of electrically conductive adhesives (ECAs) as a replacement for solder joining technology in many electronic packaging industries. It considers the fundamentals of epoxy based ECAs focusing on various categoriessuch as ICAs, ACAs and NCAs and conductive fillers incorporated within the epoxy matrix include metal fillers, carbon fillers, ceramic fillers and conducting polymers have beenreviewed. More research works in improving those performances of ICAs in replacing SnPb or Pb-free solder alloys in die attach, surface mount technologies and flip chip assemblies with electrical, mechanical and reliability enhancements. ACAs are becoming attractive in fine pitch and Pb-free interconnection areas for flat panel displays and semiconductor packaging industries.

Throughout the review it is observed that fillers incorporated into the adhesive matrix playsan important role in determining the electrical, mechanical as well as the thermal properties, of the ECAs. Among various fillers used, copper is the dominant filler used most due to its lower cost consideration in addition to its enhanced electrical properties, although it should also be pointed out that oxidation and degrading of copper filler upon aging is a major challenge on the wide usage of copper as a filler material. Replacing pure copper by alloys of copper including silver-copper alloy are discussed as a solution for degrading and oxidation properties of copper. It is also observed that metallic fillers can be replaced by carbon based conductive fillers like carbon nanotubes and graphene, where studies revealed towards the improvement of electrical properties of ECAs upon the usage of carbon based conductive fillers. Methods for modification of graphene as well as carbon nanotubes were discussed andimprovements in electrical as well as mechanical properties are evident.

It should also be noted that other than that of the filler materials used, factors such as type of adhesive resin used, processing methods selected for preparation of fillers, mixing techniques utilized, structure and shape of filler material, distribution of fillers over matrix, mixing temperature etc. will also contribute towards the resulting properties of the ECAs. While discussing about the application aspects of ECAs other than the replacement of conventional Sn-Pb soldering, today's blooming electronic technologies, like multilayer printed circuits, thin-film transistors, smart card application etc. are some of the key fields in which ECAswill be utilised in near future.

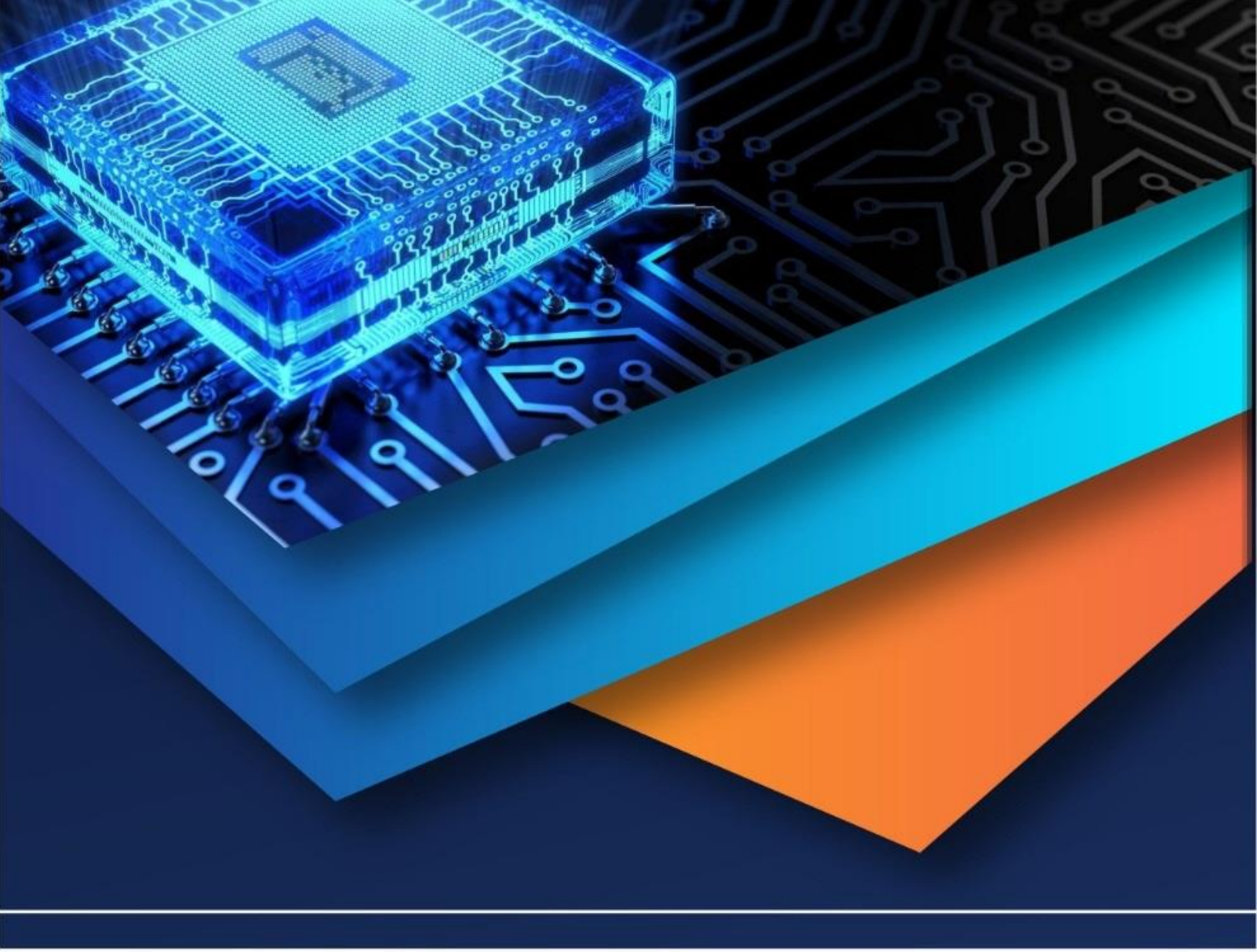
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