

Interfacial Design for Joining Technologies: An Historical Perspective

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This paper gives an historic perspective of the concept of “Interfacial Design” in joined (e.g. soldered, brazed, diffusion bonded) assemblies. During the course of history, the awareness grew that the interface in a material joint can be perceived at different length scales. With the continuing development of joining materials and technologies, it became evident that the performance of assemblies is critically dependent on the structure and composition of the multiple internal interfaces in the material joints. Resulting trends in the microstructural design of soldering, brazing, and other bonding materials by smart engineering of internal interfaces, as driven by increasingly complex technological requirements, are briefly addressed.

Keywords brazing, coatings, interfaces, joining, nanomaterials

1. Introduction

Interfaces of multi-phase materials and their assemblies (e.g., hetero-interfaces, phase boundaries, grain boundaries, and free surfaces) impose dimensional and microstructural constraints to the mobility of atoms, single defects, dislocations, electrons, photons, phonons, and plasmons. Hence, the local atom arrangement, defect structure, and coherency strain at internal interfaces are decisive for tuning the functional (e.g., mechanical, electrical, magnetic, thermal, or optical) properties of components and devices: see Ref 1-5 and references therein. Particularly in nano-structured materials, such as thin film systems and nano-composites, a relatively large volume fraction of atoms is associated with internal interfaces, which often results in “unusual” materials properties, which are different from those of the corresponding bulk material(s): e.g., an ultra-high yield strength, superparamagnetism, high catalytic activity, size-dependent optical properties, and/or a strikingly lower (pre-melting) or higher (superheating) melting point (Ref 6).

In addition, in nano-structured materials, the excess energies associated with internal interfaces often dominate the energetics

of the system: i.e., a high density of internal interfaces typically increases the total Gibbs energy of the system (Ref 7), thereby providing large driving forces for microstructural and interfacial transformations, such as interfacial segregation, wetting, complexion transition, reconstruction, intermixing and compound formation. In addition, the relatively short diffusion distances, often in combination with enhanced diffusion rates of atoms along internal interfaces, enable much faster kinetics for thermodynamic equilibration, which makes artificially, man-made nanostructured components and nano-devices very prone to degradation (Ref 8-10).

Evidently, tailoring the properties (including the chemical and mechanical stability) of internal interfaces is of cardinal importance in numerous technologies in the fields of, e.g., mechanical engineering, microelectronics, nano-photonics, catalysis, photovoltaics, and sensing devices.

In the field of traditional joining technologies (e.g., brazing, soldering, diffusion bonding), it has also long been recognized that the performance of a material joint critically depends on the microstructure of the “macroscopic” interface, as conceived by the brazed zone in between the parent joining materials. However, such a “macroscopic” perception disregards the presence of multiple internal interfaces, such as grain and phase boundaries, within the brazed zone, which actually govern the properties of the material joint. Until the present date, the concept of interfacial design (or interface engineering) at various length scales for tailoring the properties of a material joint is often still not envisioned as an integral and crucial part of joint manufacturing. Only in recent days, due to the continuing miniaturization of material components in modern nano-technologies (e.g. micro-electronics, medical implants, microelectromechanical systems and sensing devices), interfacial design is becoming a crucial task for integrating, packaging and assembling increasingly complex nano- and microscale materials and components at ever-lower temperatures (Ref 11). In particular, smart design (or engineering) of the local composition and structure of the internal interfaces in nano- and microscale systems is needed to allow faster and more reliable fabrication, continuing miniaturization, further cost reduction, and enhanced durability during service.

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With the aim to face the aforementioned technological challenge, the current authors initiated a first symposium on “Interfacial Design” within the “Joining” topic at the *Euromat 2013* conference. The initiative follows similar trends in other conference series, such as the *International Conference on Brazing, Diffusion Bonding and High Temperature Brazing*, which also organized a first session on *Nanotechnology* in 2013. Noteworthy, a new conference series on the topic of *Nano- and Microjoining* has been initiated in 2012 with a forthcoming conference in 2014 (see www.nmj2014.org). These scientific progressions in the field of joining are strongly driven by the industrial demand to control the microstructure of a material joint with its internal interfaces down to the atomic scale. In the past, the development of new joining concepts typically succeeded the first development stages of material and system design, components manufacturing, and assembly. In more recent days, with the continuous miniaturization of functional components, the joining process is becoming more and more an integrative part of the product development and manufacturing chain (Ref 12).

2. Interfaces in Material Joints: A Historical Perspective

Joining is a very ancient technology; first soldering and brazing technologies are thought to date back to before 4000 BC. Back then as nowadays, the fabrication of a good joint could only be mastered by establishing a solid bond between the two material components. A main criterion for a good joint still is the strength of the chemical bond across the macro-interface. Some of the first objects manufactured by joining technologies in ancient history, such as jewelry or weapons, have lasted until today and can be admired in archeological museums worldwide. They evidence that, already in ancient times, humans mastered the joining of all kinds of dissimilar materials, such as metals, alloys, and glasses.

For a long time, a material joint was understood as a connection of two materials, separated by a single *boundary plane* with invisible or negligible thickness and without any specific properties. Soon after the invention of different solders and brazing filler metals (alloys), the awareness grew that the properties of a material joint can be controlled by smart selection of the type of the solder or brazing filler. As a result, increasing attention was paid to the resulting microstructure of the joined zone between the two base materials. From this time on, the microstructure and constituents of the macroscopic interface were studied in more detail. In the case of a brazed joint, the macro-interface of the joint involves a zone with a thickness of a few tenths of a millimeter, as constituted by the reacted brazing filler (the modified brazing filler alloy) in the center and its reaction zones with the base materials. Similar concepts were considered for material joints produced by other metallurgical bonding processes, such as soldering or diffusion bonding processes. Nevertheless, it was (and still is) often naively assumed that the interface, as defined by the brazing zone, possesses similar properties as those of the applied brazing filler. Furthermore, the quality of a joint was (and still is) often simply judged on the basis of the size and shape of visible defects and discontinuities in the brazing zone. Hence the procedure for the optimization of a given joining process



Fig. 1 An exemplary metal-ceramic joint: the “macro-interface” of the joint involves the zone with a thickness of a few tenths of a millimeter, as constituted by the reacted brazing filler (the modified brazing filler alloy) in the center and its reaction zones with the base materials. For metal-ceramic joints, the two “micro-interfaces” as associated with the opposite reaction zones have contrasting chemical bonding characteristics

often still proceeds by a series of trial-and-error joining experiments with a pre-selection of different brazing filler materials.

With the development of joints of dissimilar materials with very different electronic bonding configurations, such as metals and ceramics, the brazing community recognized that it is not sufficient to optimize only the applied brazing filler alloy and its resulting microstructure after joining. To optimize the properties of the material joint it became more and more essential to control the contrasting chemical reactions at the opposing micro-interfaces of the braze with the dissimilar base materials (see Fig. 1), as tailored by the addition of so-called (re-)active elements (e.g., Ti, Cr, and Zr).

However, the quality of a brazed metal-ceramic joint not only relies on strong chemical bonding at the opposing microinterfaces and, consequently, a good wetting behavior of the brazing filler with the ceramic (Ref 13-16). The joint quality also critically depends on the residual stress levels in the brazed zone, as originating from the characteristically large difference in thermal expansion coefficients between the metal and the ceramic (Ref 17-21). Ideally the brazing filler should have a high plasticity and an intermediate (or even gradient) thermal expansion coefficient between the metal and the ceramic to prevent large stress gradients in the joint zone (Ref 21-24). Unfortunately, such ideal brazing fillers are not always available and then complex interface engineering is needed to deal with the large thermal expansion mismatch between the two base materials. Typical strategies to reduce the stress gradient in the metal-ceramic joint zone are the application of: (i) a combination of one or more brazing fillers with a reactive metallization layer(s) on the ceramic base material (Ref 25-28), (ii) a succession of interlayers with different thermal expansion coefficients to more gradually bridge the jump in thermal expansion coefficient between the metal and the ceramic base components (Ref 18, 29-31), (iii) composite brazing fillers (Ref 32-39).

Ceramic-particle-reinforced active brazing fillers are a typical example of the above mentioned composite brazing fillers for active brazing of metals to ceramics (Ref 17, 32, 40), see Fig. 2. Due to the increasing number of internal metal-ceramic interfaces induced upon addition of (micro-sized) ceramic particles to the active brazing filler (to tailor both the

thermal expansion coefficient and the mechanical properties of the braze), high-quality joints can only be achieved by increasing the concentration of the active element (here: Ti) in the brazing filler with respect to the optimum content in the unreinforced brazing filler (Ref 17, 22, 41). Namely, the active element is needed to establish firm chemical bonding at all internal metal-ceramic interfaces, i.e., between the brazing filler and ceramic base material, as well as between the ceramic particles and the brazing filler matrix.

Also for metal-metal joints, such as lead-free soldering of Cu interconnects, it can be beneficial to add nano-sized metallic particles to the solder to improve the creep resistance (Ref 42-44) and to optimize the thermo-mechanical fatigue behavior of the interconnections (Ref 45, 46). For example, if Cu nanoparticles are added to a Sn-Ag-Cu solder when joining Cu components, the growth of the brittle Cu_6Sn_5 intermetallic

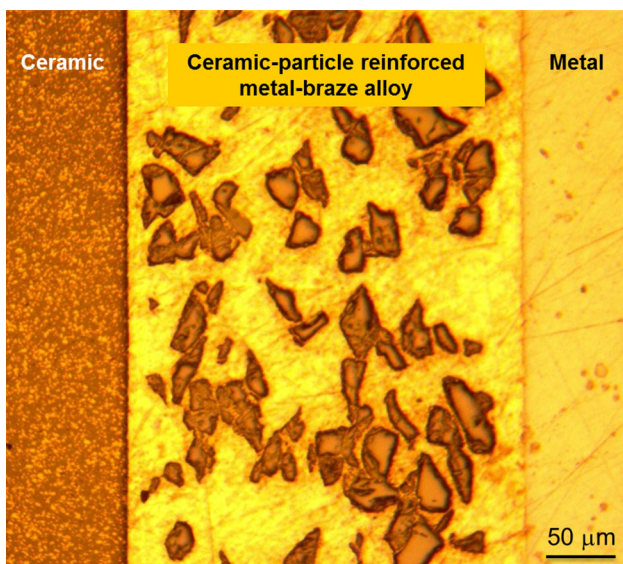


Fig. 2 Metal-ceramic joint (left $\text{Si}_3\text{N}_4/\text{TiN}$ ceramics, right 14NiCr14 steel), as produced by the application of a ceramic-particle-reinforced active brazing filler (center AgCuInTi reinforced with SiC particles)

phase during joint service can be suppressed by the formation of the Cu_6Sn_5 around the Cu nanoparticles, thus depleting Sn from the competing reaction with the Cu base material (Ref 45).

Another recent development in interfacial design for joining technologies is the research on the application of metallic nanomultilayers (NMLs) systems, e.g., reactive nanofoils as a potential local heat-source for the joining of heat-sensitive nano-structured materials and micro-scale devices (Ref 47-56), as applied in, e.g., microelectronics, packaging, sensors, and implantable medical devices. Reactive NMLs foils or coatings, constituted of alternating Ni-Al, Ti-Al, and Pd-Al sublayers with a thickness in the range of 3-100 nm, as prepared by conventional magnetron sputtering techniques, are among the most promising systems for such benign joining technologies. Thermal activation of the self-propagating chain of exothermic reactions associated with intermetallic phase formation in the NMLs results in the very fast release of thermal energy. Application of reactive NML foils or coatings between two base components thus enables instantaneous release of local heat in the vicinity of interface between the contacted base materials to establish a metallurgical joint, thereby restricting the heat-affected zone to a few hundreds of micrometers from the joint line.

Only very recently, a novel design of nanostructured brazing filler materials was proposed (Ref 57-70), which exploits nano-effects, like fast interfacial reaction kinetics, grain boundary wetting, and melting point depression by nano-confinement to realize brazing at reduced temperatures (as compared to the brazing temperature for the corresponding bulk brazing alloy). Representative examples of such joining materials are brazing pastes from nanoparticles with an intelligent, temporary coating (Ref 71, 72), and nano-multilayers, constituted of alternating nanometer-thick layers of a brazing filler metal and a chemically inert barrier (e.g., a nitride, oxide or refractory metal) (Ref 69, 70) see Fig. 3. Evidently, such nano-effects are controlled by the local structure, chemical composition, and properties of the different internal interfaces. Hence interfacial design is a crucial first step in the development and application of such novel nano-structured joining materials. For example, the crucial role of the structure of internal interfaces on melting behavior was demonstrated for an In nano-particle in an Al

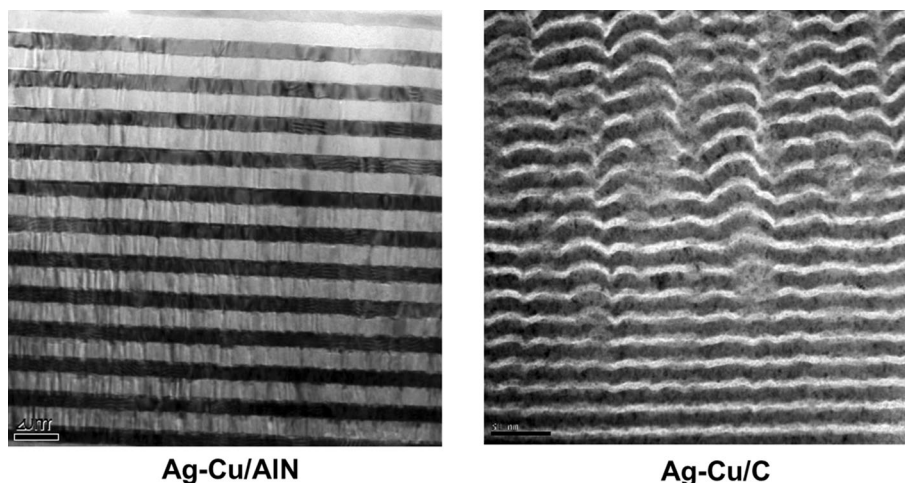


Fig. 3 Nanomultilayers for joining: multiple internal interfaces between nanometer-thick layers of a typical braze alloy (Ag-Cu; dark) and a chemically inert barrier layer (AlN, C; bright)

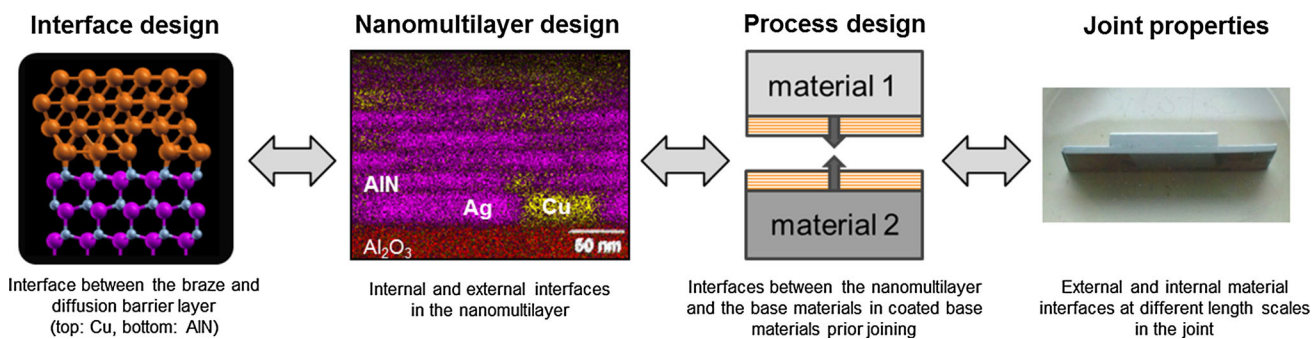


Fig. 4 Illustration of the complex interplay between competing reactions at neighboring interfaces in nanostructured brazing fillers in a multilayer configuration

matrix; a coherent In-Al interface resulted in superheating, whereas a melting point depression occurred for the non-coherent In-Al interface (Ref 73). On this foundation (i.e., by smart interfacial design), strikingly large melting point depressions and very fast reaction kinetics can principally be achieved in a nanomultilayer configuration (Ref 70).

Some of the challenges which joining technologies are facing today (e.g., joining at low temperatures, while assuring operation of the joined assemblies at much higher temperatures) can only be mastered by exploring such new nano-scale-based concepts. Hence, a complex design of brazing filler metals and solders with a high density of smartly engineered internal interfaces is needed. Each internal interface will contribute to some extent to the physical and chemical properties (e.g., mechanic, thermal, electronic) of the material joint and hence principally should be tailored (“designed”) to optimize and/or functionalize the joint assembly. Unfortunately, interfacial design down to the atomic scale is far from trivial, particularly due to the difficulties in controlling the kinetics of competing reactions (e.g., interdiffusion, intermixing, heterogeneous nucleation, and growth) at neighboring interfaces, as well as the developing stress gradients upon heating. For example, for the joining of base materials coated with nano-multilayers, the interface reactions in the nano-sized brazing filler (e.g., alloying, phase separation), those between the brazing filler and the barrier layer, as well as those between the brazing filler and the base materials, have to be controlled, see Fig. 4.

3. Conclusions

The concept of “Interface Design” in joining technologies has evolved during centuries and is still under discussion. During the course of history, the awareness grew that the interface in a material joint can be perceived at different length scales. With the continuing miniaturization of engineered components, it became more and more evident that the performance of material assemblies is critically dependent on the structure and composition of the multiple internal interfaces in the joint. Nowadays, a complex design of brazing filler metals and solders with a high density of smartly engineered internal interfaces is often needed to meet industrial requirements. The various interfaces in nanostructured joining materials offer us a new degree of freedom for optimization of the joining process. Each internal interface contributes to some extent to the physical and chemical properties (e.g., mechanic,

thermal, electronic) of the material joint and therefore should be tailored (“designed”) to optimize and/or functionalize the joint assembly. Unfortunately, such interface design down to the atomic scale is still far from trivial, particularly due to the difficulties in controlling the kinetics of competing reactions at neighboring interfaces and the developing stress gradients upon heating. The joining community has to face this big challenge. With this first symposium on *Interface Design in Joining*, controlling the structure and properties of interfaces in the joint should more and more become an integral part of advanced joining technologies.

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