Journal of Materials Science & Technology 34 (2018) 192–197

Contents lists available at ScienceDirect

Journal of Materials Science & Technology

journal homepage: www.jmst.org

Direct joining of oxygen-free copper and carbon-fiber-reinforced plastic by friction lap joining

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ARTICLE INFO

Article history:
Received 3 April 2017
Received in revised form 25 April 2017
Accepted 29 April 2017
Available online 2 November 2017

Keywords:
Friction lap joining
Metal
Plastic
Carbon-fiber reinforced thermoplastic
Dissimilar materials joining

ABSTRACT

Oxygen-free copper (Cu) was successfully joined to carbon-fiber-reinforced thermoplastic (CFRTP, polyamide 6 with 20 wt% carbon fiber addition) by friction lap joining (FLJ) at joining speeds of 200–1600 mm/min with a constant rotation rate of 1500 rpm and a nominal plunge depth of 0.9 mm. It is the first time to report the joining of CFRTP to Cu by FLJ. As the joining speed increased, the tensile shear force (TSF) of joints increased first, and decreased thereafter. The maximum TSF could reach 2.3 kN (15 mm in width). Hydrogen bonding formed between the amide group of CFRTP and the thin Cu_2O layer on the Cu surface, which mainly contributed to the joint bonding. The influence factors of the TSF of the joints at different joining speeds were discussed. The TSF was mainly affected by the joining area, the degradation of the plastic matrix and the number and the size of bubbles. As the joining speed increased, the influence factors varied as follows: the joining area increased first and then decreased; the degradation of the plastic matrix and the number and the size of bubbles decreased. The maximum TSF was the comprehensive result of the relatively large joining area, small degradation of the plastic matrix and small number and sizes of bubbles.

1. Introduction

Nowadays, carbon-fiber-reinforced plastic (CFRP) materials have been increasingly used as structural materials in aerospace, automobile, and electronic industries for a reduction of fuel consumption, because of their light weight, high specific strength and very low corrosion rate [1]. Among CFRP materials, carbon-fiber-reinforced thermoplastic (CFRTP) materials, which are highly processable, especially draw lots of attentions. However, the inferior thermal and electrical conductivities of CFRTP materials have largely limited their application. As we know, metals are characterized by the high thermal and electrical conductivity and superior specific strength [1–3]. Therefore, the hybrid joining of plastics including CFRTP and metals are highly demanded for the structural applications where plastics and metals can compensate each other for various advantages to achieve a more flexible structural design, high performance, cost saving, etc [4–6].

However, it is not easy to join plastics and metals since there is a huge difference on physical and chemical properties. Adhesive bonding and mechanical fastening have been commonly reported to apply for the joining of plastics to metals, and relatively strong hybrid joints could be obtained [7]. However, some drawbacks always exist for these conventional joining methods. For example, adhesive bonding is always involved with environmental issues and inferior long-term stability, and mechanical fastening are usually associated with inflexible design challenges and stress concentrations [7]. To solve these problems, researchers have been dedicating on the exploring of some novel joining techniques, such as laser welding [2,8,9], friction stir spot welding [6,10], and ultrasonic welding [3,11].

Katayama and his co-workers [2,8,12,13] have made some good reports on the laser welding of different plastics including CFRP to metals such as steel and Al alloys. They found that the joints for CFRP to different metals could all achieve a high tensile shear force (TSF) of more than 3 kN (20 mm in width) by the chemical or physical bonding on atomic, molecular or nano-sized level between the melting plastic and the oxide of the metal surface, as well as the mechanical bonding. During laser welding, the large pressure produced by the expansion of bubbles was benefit for the bonding of plastics with metals. Therefore, laser welding shows a great potential for the joining of plastics and metals, however, most of previous investigations have focused on the laser welding of plastics to Al, Mg, Ti and steel. So far, there are only limited preliminary papers on the joining of plastic to Cu.

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https://doi.org/10.1016/j.jmst.2017.10.019
1005-0302/

It is well known that Cu is widely used in electronic components and air-conditioning condenser pipes. The joining of Cu to polymer is of great significance since the Cu-polymer hybrid joints can reduce the weight of the components purely made of Cu, which reduce the energy-consuming, thereby reducing the products costs. Different from that strong laser welds of plastic to Al or steel were usually obtained, not so strong plastic-Cu joints were produced by direct laser welding and also the energy input largely increased [14]. Tamrin et al. [7] pointed out that the feasibility of laser welding was largely dependent on the laser absorptivity of materials. It is much more difficult for the joining of plastics to Cu materials compared to Al and steel, since Cu has high reflectivity and conductivity. Therefore, other joining methods are needed to be applied to join plastics and Cu materials. Although the joining of plastics and metals has been also found to be feasible by friction stir spot welding or ultrasonic welding [3,6,11,15], yet, dimensions and geometries of the joints are limited. Besides, so far, no results on the joining of plastics to Cu materials using friction stir spot welding or ultrasonic welding have been reported.

In order to solve the shortage of all these joining methods of the joining of plastics to Cu materials, friction lap joining (FLJ) seems to be a potential one. FLJ, a new variation of friction stir welding (FSW) which is widely applied in various metals and composites [16,17], has been recently developed for the joining of plastics and metals [1,18,19]. The main difference between FLJ and FSW is that during FSW, a stir pin attached to a tool is usually used to assist the material flowing, while no stir pin for FLJ. FLJ mainly involves that friction heat between metals and tool is transferred into plastics, resulting in the melting of plastics near the interface, and then the joining of plastics and metals is achieved under the pressure of the tool and clamping apparatus after solidification of plastics [1,18].

Now FLJ has been reported to successfully join plastics and metals such as Al, Mg alloys and steel [1,18,19]. For example, MC nylon-6 could directly join with Al and Mg alloys by FLJ [18,19]. It was also found that similar to that in laser welding, bubbles could be observed in the joints, and strong FLW joints was obtained when the area fraction of bubbles was less than 8% via welding process optimization [19]. Besides, during direct FLJ of CFRTP and 5052 Al alloy, hydrogen bonding formed between the plastic matrix and an oxide of 5052 Al sheet surface, which contributed mainly to the joint bonding [1]. Furthermore, it was found that by a chemical surface treatment (i.e. silane coupling) on the Al sheet surface before FLJ, the tensile shear strength of the joint could even be over 20 MPa and an efficiency of the joint of 97% could be obtained with the joint fracturing at the base material of CFRTP [20]. Therefore, FLJ shows a big potential for the joining of plastics to Al, Mg alloys and steel, however, the feasibility of the FLJ of plastics to Cu materials still remains unknown.

Since the joining of plastics and metals are now still in the developing stage, in order to enlarge the knowledge of the plastic-metal hybrid joining, the joining of CFRTP to Cu was conducted by FLJ at different joining speeds in this study. The objective is to explore the feasibility of directly joining of plastics to Cu materials, to evaluate the joint characteristics, and to clarify the influence of the joining speed on the tensile shear properties of the joints.

2. Materials and experimental procedure

The as-received materials were 3-mm-thick CRFTP sheets (polyamide 6 (PA6) with 20 wt% carbon fiber addition) made by injection molding, and 2-mm-thick oxygen-free copper sheets. The diameter and length of the carbon fibers were 10 μm and about 500 μm, respectively. The average tensile strengths of the CFRTP were 140 MPa in the flow direction and 117 MPa in the transverse direction, and about more details on the CFRTP, please refer to the previous work [1]. Before FLJ, the Cu sheets were ground in the flowing water with #800 emery paper, and CFRTP sheets were dry-ground with #80 and #800 emery paper. The CFRTP sheets were friction lap joined to Cu at joining speeds of 200–1600 mm/min with a constant rotation rate of 1500 rpm. A tool plunge depth of 0.9 mm, a tilt angle of 3 degree, and a lap width of 30 mm was used by a steel tool with a shoulder diameter of 15 mm without a stir pin. For the temperature measurement during FLJ, a K-type thermocouple was inserted at Cu sheet/CFRTP sheet interface at the center of the joined area.

The specimens for microstructural observation were first cut perpendicular to the joining direction, mounted in epoxy resin, and ground and polished with silica solution. The microstructural observation of these specimens was then performed via optical microscopy (OM) and transmission electron microscopy (TEM). To test the TSF, specimens were cut perpendicular to the joining direction with a width of 15 mm. Tensile shear tests were carried out in a regular tensile machine at the crosshead speed of 0.5 mm/min. For each joining condition, three tensile specimens were tested, and in order to reduce the effect of travel position on the microstructure and mechanical properties, the specimens were all cut from the travel distance of 60–120 mm. The fracture surfaces of the tensile shear specimens were observed using OM, SEM with energy dispersive X-ray spectroscopy (EDS). The residual CFRTP areas on the fractured surface of Cu were measured manually using Photoshop software.

3. Results and discussion

After FLJ, Cu could successfully join with CFRTP at all the joining speeds, and the typical surface morphologies of FLJ joints of Cu to CFRTP at 200 and 1600 mm/min are shown in Fig. 1. The advance side (AS) and retreating side (RS) were located at the CFRTP side and Cu side, respectively. At all the joining speeds, the joints could not be separated apart by human hand force, which suggested that CFRTP should have joined well with Cu at all the joining speeds.

The variation of the TSF of the CFRTP-Cu FLJ sheets with the joining speed is shown in Fig. 2. It was obvious that as the joining speed increased, the TSF increased first, and then decreased. The TSF of the joints achieved the maximum of 2.3 kN at 600 and 800 mm/min at a nominal plunge depth of 0.9 mm. Therefore, it is feasible to join CFRTP to Cu directly by FLJ.

In order to explain the variation trend of TSF with the joining speed, the temperature profiles at the joint center line at different joining speeds were measured, as shown in Fig. 3. At each joining speed of 200 to 1600 mm/min, the maximum temperature was all over the thermal decomposition temperature (350 °C) and the

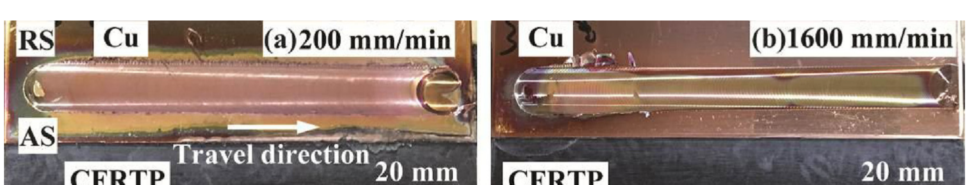

Fig. 1. Typical marostructural morphologies of friction lap joints of CFRTP to Cu at (a) 200 and (b) 1600 mm/min.

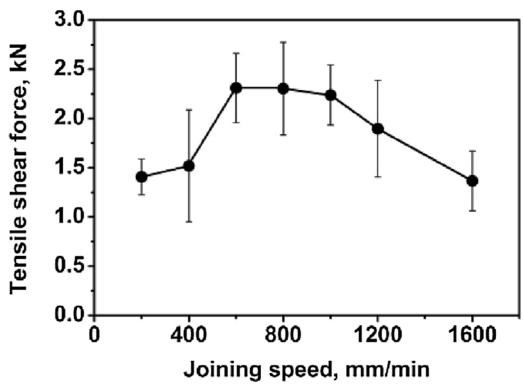

Fig. 2. Variation of tensile shear force with joining speed of CFRTP/Cu friction lap joints.

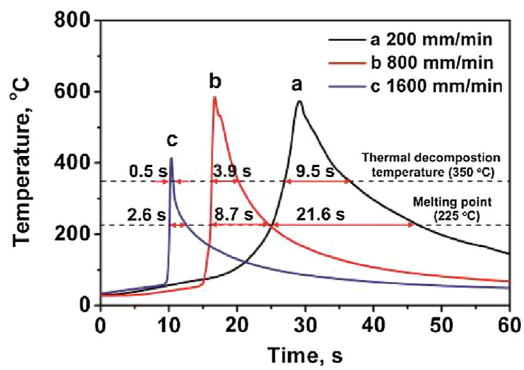

Fig. 3. Temperature measurement of CFRTP-Cu joints at different joining speeds of 200, 800 and 1600 mm/min during FLJ.

melting point (225 °C) of PA6 (the matrix of CFRTP). As the joining speed increased from 200 to 1600 mm/min, the holding time above the melting point and thermal decomposition temperature decreased, from 21.6 s into 2.6s, and from 9.5 s into 0.5s, respectively. Therefore, during FLJ, plastic at all the joining speed should have melted and decomposed in some degree depending on the high temperature and holding time.

The typical macrostructural cross sections of the Cu-CFRTP FLJ joints are shown in Fig. 4. It was obvious that the interface of each joint showed a concave shape resulted from the downward force of the welding tool. The curvature of the interface, depending on the melting and softening of CFRTP, decreased with the joining speed. A white melted zone (marked by black dot lines in Fig. 4) was observed in the CFRTP near the interface of each joint. As the joining speed increased, the melted zone became thinner, and at 1600 mm/min, the white melted zone was even hardly found expect for a localized one at the edge of AS, as marked by the ellipse. This should be related to the shorter holding time for melting at high temperature for the higher joining speed (Fig. 3).

The magnified optical images of the joints in the center at different joining speeds are shown in Fig. 5. It indicated that in these melted (or re-solidified) zones, there were a large number of bubbles. These bubbles were reported to mainly come from the thermal decomposition of plastic, although a small part might also came from the air and water vapor [1,2]. As the joining speed increased, the fraction and the size of bubbles decreased (Fig. 5). Besides, a number of carbon fibers gathered in some regions near the interface of Cu and CFRTP, while in some other regions the number of carbon fibers reduced, even no carbon fibers existing (e.g. Fig. 5b). This phenomenon was mainly attributed to the different flowing rates for the plastic matrix and carbon fibers. It was obvious that the distribution of carbon fibers changed more for the lower joining speed.

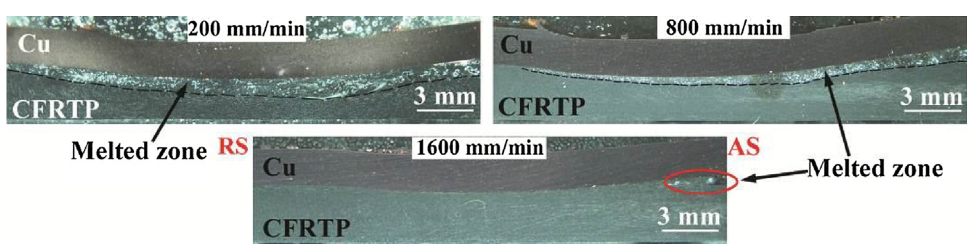

Fig. 4. Typical macrostructural cross-sections of FLJ joints of CFRTP-Cu at different joining speed.

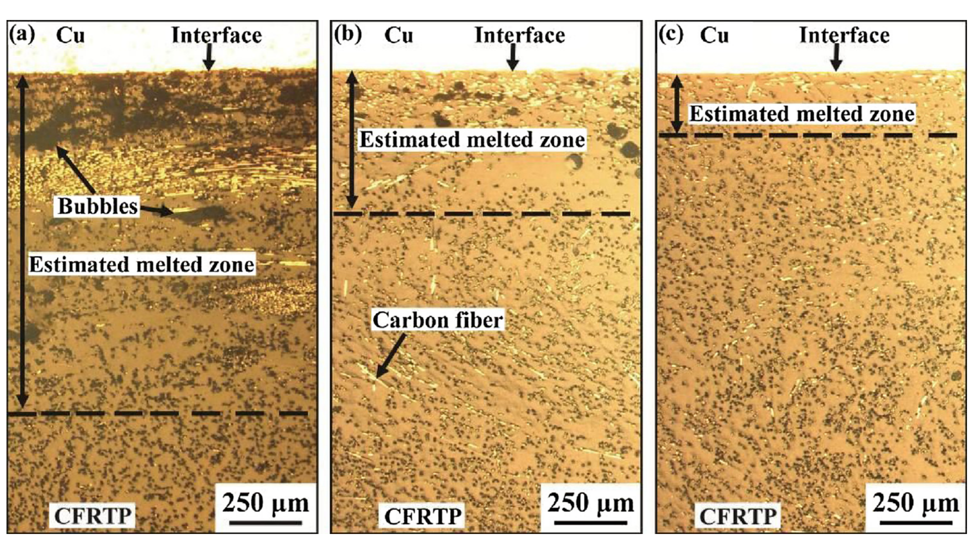

Fig. 5. Typical microstructure of the interface on cross-sections of FLJ joints of CFRTP-Cu at different joining speed: (a) 200, (b) 800, and (c) 1600 mm/min.

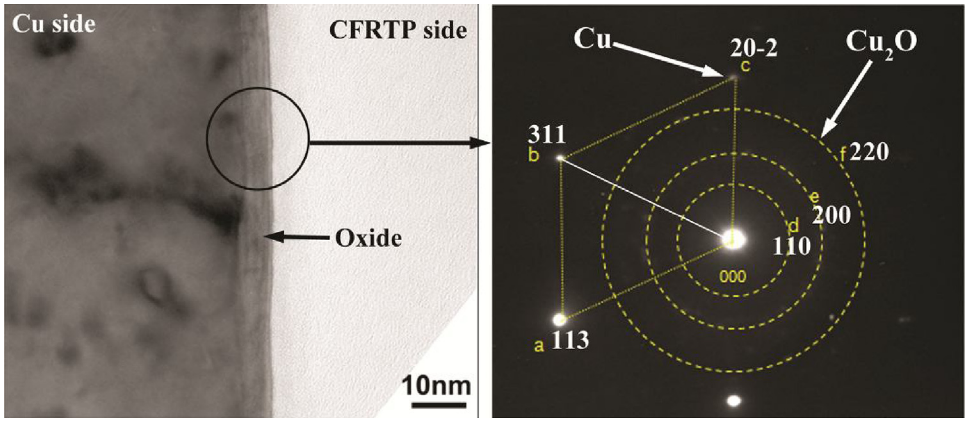

Fig. 6. Typical TEM and selected area diffraction (SAD) of Cu-CFRTP joint interface showing Cu_2O thin transition layer.

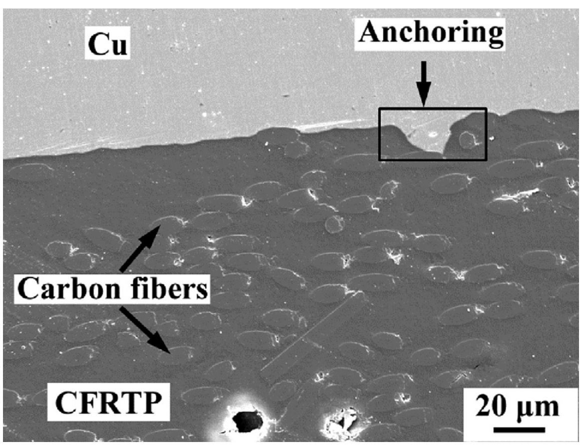

Fig. 7. Typical SEM image of CFRTP-Cu joint interface at 800 mm/min showing anchoring effect.

In this study, the change of the orientation and distribution for carbon fibers was regarded as the rough definition criteria for the estimated melted zone. The thickness of the melted zone decreased from about 1150 μm to only 210 μm when the joining speed increased from 200 mm/min to 1600 mm/min (Fig. 5). A thinner melted zone with less bubbles for a higher joining speed should be the result of the reduced holding time for melting and thermal decomposition of plastic.

Fig. 6 shows the typical TEM image and selected area diffraction (SAD) of the CFRTP-Cu joint at 600 mm/min. It was found that Cu transited into CFRTP continuously without any void or gap. There was a very thin Cu_2O transition layer (confirmed by the SAD in Fig. 6) at the interface of Cu and CFRTP, which suggested that Cu_2O directly joined with PA6 on the nanoscale level. As we know, an amide group (CONH) in PA6 is easy to form a hydrogen bonding with the oxide on the metal surface [1]. Therefore, in this study, hydrogen bonding between PA6 and Cu_2O on the Cu surface should have formed.

In addition, some rough surface morphology, such as anchors, contributed to increase the interface joining area, and a typical example is shown in Fig. 7. However, this kind of anchor with a small depth and width was just observed in some local zones, and the contribution of the anchoring effect to the TSF was very limited. It is well-known that plastics joined with metals usually by chemical or physical bonding, or mechanical interlocking effect [2]. For FLJ of plastic to Al alloys and steel, it was reported that hydrogen bonding contributed the most to the joint bonding [1,21]. Therefore, in this study, the strong joining between CFRTP and Cu without any special surface treatment should be also mainly attributed to hydrogen bonding (Fig. 6).

The typical tensile shear fracture surfaces of the Cu/CFRTP FLJ joints are shown in Fig. 8. On the fractured Cu side at all joining speeds, two regions including a naked Cu surface and a surface with some residual materials existed. The area of the residual materials at 800 mm/min was larger than that for 200 and 1600 mm/min. These residual materials might be CFRTP. On the CFRTP side, a number of small bubbles were observed. In order to confirm the residual materials, SEM-EDS of the Cu fracture surfaces was detected, and the typical SEM-EDS of the Cu fracture surface of the joint at 800 mm/min is shown in Fig. 9.

SEM-EDS analysis showed that the residual materials contained two types of morphologies with carbon element (marked as 1 and 2 in Fig. 9), suggesting that the residual material on the Cu surface was indeed plastic with carbon fiber, i.e. CFRTP. The first one type of residual CFRTP on the fracture surface of Cu side (marked as 1 in Fig. 9) exhibited an irregular shape with deformation characteristics, suggesting these CFRTP must have deformed during tensile shear test. It indicated that Cu must have joined with CFRTP by a strong joining bonding (hydrogen bonding in this study) so that the deformation took place in the CFRTP side near the interface. The second one showed a circle or ellipse shapes just like the bubble shapes (marked as 2 in Fig. 9), which suggested that the bubbles might have acted as the fracture sites.

From the fracture surface morphologies (Figs. Fig. 8 and 9), it suggested that the fracture of the CFRTP-Cu joints occurred predominantly by the mixture of adhesive (at the interface between Cu and re-solidified plastic layer) and cohesive (within the re-solidified CFRTP layer) failures [22]. Actually in this study, three regions acted as the fracture sites, and they were the unjoined interface of the CFRTP and Cu, the CFRTP in the re-solidified zone and the bubbles. The schematic showing estimated fracture pass during tensile shear test is shown in Fig. 10. As we know, the fracture would occur along the weakest region of the joint. When there was no joining or the joint bonding was weak enough, the fracture would preferentially occur along the interface of Cu and CFRTP, i.e. adhesive failure. Once there was a strong joining (it is hydrogen bonding in this study), fracture would occur along the re-solidified CFRTP or bubbles. In other words, the residual CFRTP area on the Cu fracture surface could roughly represent the joining area of the CFRTP-Cu joints.

As we know, the TSF of the joints is determined by both the joining area and the joint strength. The larger the joining area and the joint strength are, the larger the TSF of the joint is. Therefore,

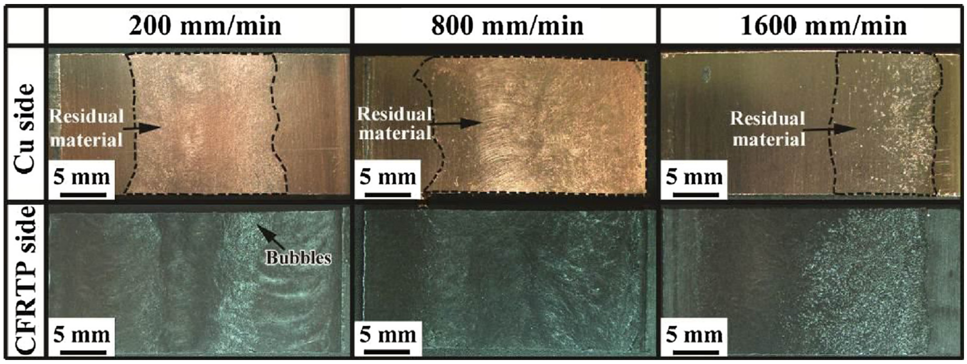

Fig. 8. Typical macrostructures of the opposing fractured surface of CFRTP-Cu FLJ joints after tensile shear test.

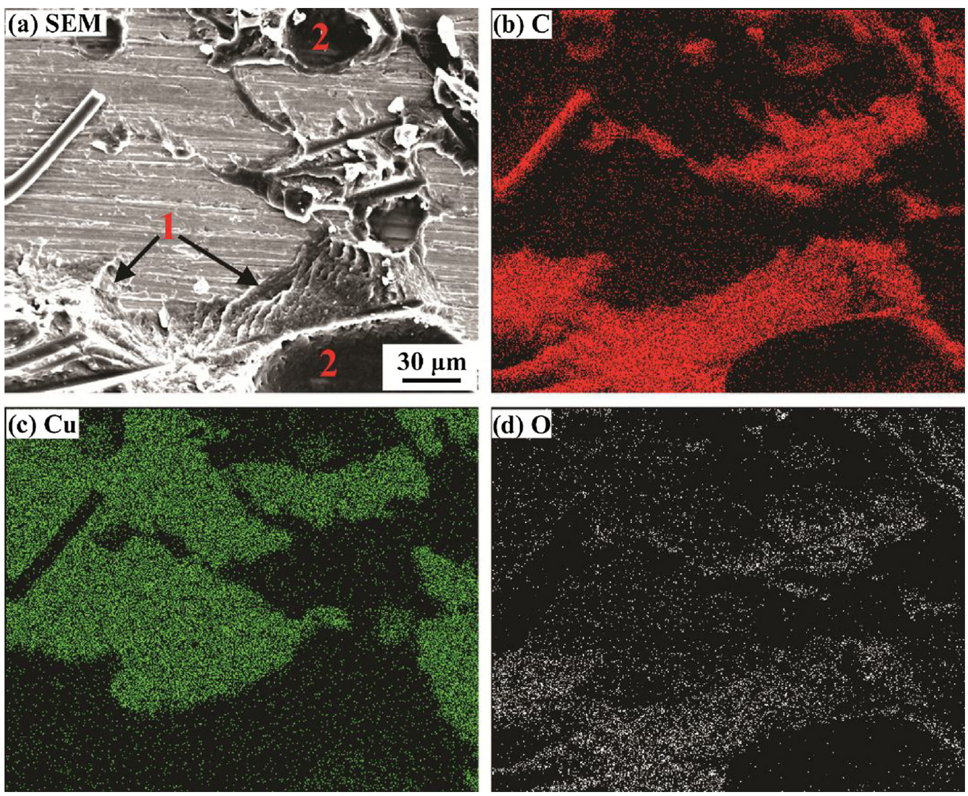

Fig. 9. Secondary electron images (a) and elements distributions by EDS on the Cu side of fractured Cu/CFRTP joint at 1500 rpm and 800 mm/min: (b) C, (c) Cu and (d) O elements.

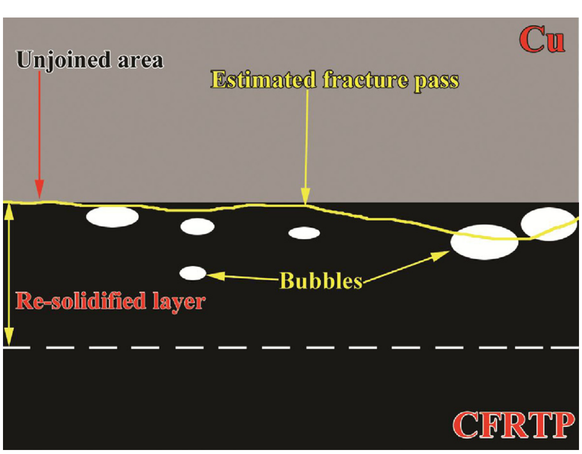

Fig. 10. Schematic of estimated fracture pass of Cu/CFRTP FLJ joints.

the TSF variation trend will be explained by the trend of both the joining area and joint strength as follows:

A. The joining area

The residual CFRTP area (representing the joining area) was measured, as shown in Fig. 11. It showed that the residual CFRTP area increased first, and then decreased with the joining speed. The maximum residual CFRTP area was achieved at 1000 mm/min. Therefore, the joining area also showed the similar trend with the joining speed. Actually, the joining area was also enhanced by the curvature of the interface by the increment of the practical fracture pass length. The curvature of the interface of CFRTP-Cu joint decreased as the joining speed increased. It means the practical fracture pass length increased more for those joints with lower joining speeds.

It should be pointed out that the error bar of the TSF and residual CFRTP area especially at 400 mm/min (Figs. 2 and 11) was not small. This phenomenon should be attributed to the fact that Cu has a very high thermal conductivity, which made it difficult to reach

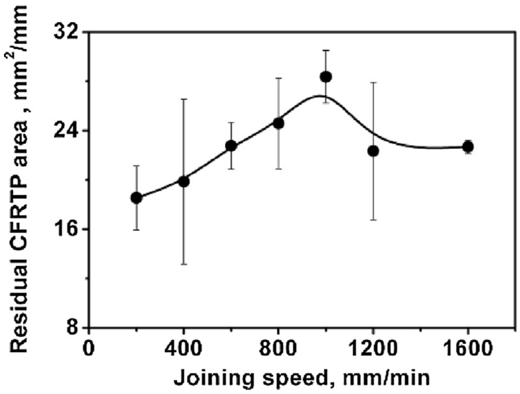

Fig. 11. Measured residual CFRTP area on the Cu fracture surface with joining speed.

heat equilibrium at some joining speeds during FLJ, resulting in different joining widths at different travel positions. As a result, the joints showed different joining areas and TSF at different travel position, which lead to the scattering data.

B. The joint strength

According to the analysis above, in the joining zone, the joint fracture would occur along the re-solidified CFRTP and/or bubbles (Fig. 10). Therefore, the joint strength was largely affected by the strength of re-solidified CFRTP and the number and size of bubbles. It was reported that the thermal decomposition of the PA6 matrix during FLJ process resulted in the decrement of its strength, as the measurement of the weight-averaged molecular weight [1]. A higher temperature and longer holding time lead to a larger decrement of the strength of the PA6 matrix. This means that the strength of the re-solidified PA6 matrix increased as the joining speed increased. Besides, for CFRTP, the change of carbon fiber distribution and orientation during FLJ reduced their strengthen effect (Fig. 5), thereby further decreased the strength of the re-solidified CFRTP. As the joining speed increased, the loss of the strengthen effect of the carbon fibers decreased.

For bubbles, it was reported to have a complex influence on the joint strength during laser welding [2]. On one hand, the formation of bubbles was benefit for the joining of metal and plastic, because the expansion of bubbles would produce a large pressure and push the melting plastic into the metal. On the other hand, bubbles themselves might act as fracture pass sites, which reduced the joint strength. Only the bubbles with small size were benefit for the joint strength [7,23]. However, unlike the case during laser welding that the bubbles almost remained in the same position where it formed, the bubbles during FLJ could flow out accompanying the melted plastic since there was a large downward force on the joint. It was reported that the bubbles remaining in the re-solidified plastic zone reduced the FLJ joint strength [19].

In this study, the too slow or too fast joining speeds were not benefit for the formation of a strong joint at the nominal plunge depth of 0.9 mm. For the too slow joining speed (e.g. 200 mm/min), the PA6 was largely thermal decomposed, which resulted in the formation of numbers of large bubbles and the large decrement of the strength of the re-solidified plastic. For the too fast joining speed (e.g. 1600 mm/min), the joint cooled down rapidly (Fig. 3). As a result, there was not enough time for the formation of hydrogen bonding, leading to a small joining area, and thus a small TSF exhibited. For the medium joining speeds (e.g. 600 and 800 mm/min), however, large joining areas with a relatively large interface curvature exhibited (Figs. Fig. 4 and 11). In addition, the degradation of the plastic matrix, and the number and sizes of bubbles were relatively small as the result of relatively short holding time at high temperature. All of these resulted in the maximum TSF at 600 and 800 mm/min at the nominal plunge depth of 0.9 mm.

In this study, FLJ produced sound Cu-CFRTP joints with relatively high strength, showing the feasibility of the joining of plastics to Cu materials. FLJ probably shows a larger advantage over laser welding in the joining of plastics to Cu materials because of its much lower energy input and the capacity to easily produce plastic-Cu joints with high strength. Therefore, based on these advantages, FLJ might become a very important joining method for plastics to Cu materials in the future.

4. Conclusion

The lap joint of oxygen-free copper and CFRTP was successfully made by friction lap joining without any special surface treatment and adhesives. Hydrogen bonding formed between the amide group of CFRTP and Cu_2O on the Cu surface, which mainly contributed to the joint bonding. As the joining speed increased, the TSF increased first, decreased thereafter, and the maximum TSF could reach 2.3 kN. The TSF was mainly affected by the joining area, the degradation of the plastic matrix and the number and the size of bubbles. As the joining speed increased, the joining area increased first and then decreased; the degradation of the plastic matrix and the number and the size of bubbles decreased. The maximum TSF was the comprehensive result of the relatively large joining area, small degradation of the plastic matrix and small number and sizes of bubbles.

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