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Surface Modification of Recycled PET Plate by Particle Implantation and Deposition with Plasma Spraying†

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Abstract

The polyethylene terephtalate (PET) is widely utilized for high performance as a food and beverage container due to its excellent mechanical and chemical properties. The consumption of PET material is expected to increase more rapidly. Consequently, the recycling of waste PET is urgently needed to reduce environmental problems and economic costs. The purpose of this research is to endow waste PET materials with a new function by spraying of metal and ceramics such as Cu and TiO_2. The recycled PET plate substrate for plasma spraying was prepared from waste PET bottles. It is found that Cu and TiO_2 powder could be sprayed on the surface of the recycled PET plate without heat damage and transformation of the substrate. In specific spray conditions, the implantation of melted Cu and TiO_2 particles, which retained their original shape, into the PET substrate was also observed and this is an unusual phenomenon in plasma spraying. In this research, the possibility of production of functional PET plates with electric conductivity and wear resistance was found by controlling the plasma spray conditions.

KEY WORDS: (Plasma spray) (Cu powder) (TiO_2 powder) (Recycling) (PET substrate) (Particle implantation)

1. Introduction

PET has superior characteristic such as high melting and glass transition temperatures and a glass-like transparency. Due to these characteristics, PET is widely utilized for bottles of soft drinks, wine, beers and food packing. The consumption of PET bottles increases year after year and the waste PET bottles cause environmental problems and the consumption of energy and natural resources. Therefore, the recycling of waste PET bottle is urgently needed to reduce environmental problems and economic costs. The application of plastic, such as waste PET bottles to thermal spraying for the recycling is done by producing a plastic spray powder for providing corrosion resistance on the metal substrate.

PET has a wide range of mechanical properties attainable by variations of its molecular weight, orientation and crystallinity. However, such properties as heat, wear resistance and electrical conductivity are inferior compared with other materials. In order to improve such properties, surface modifications, conferring new functions, is being tried using waste PET bottles. The present study examines the coating formation for providing the recycled PET plate with a new function by a plasma spraying technique which make it possible to attain a thick coating of various material in short time. The flow of this research is shown in **Fig.1**.

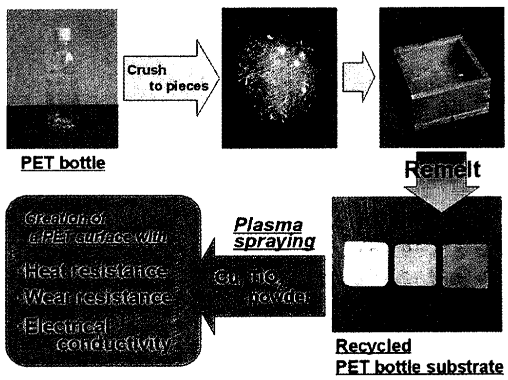

Fig.1 Flow of this research

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2. Materials and experimental procedures

2.1 Materials

Cu powders and rutile TiO_2 powders were used for plasma spraying. Recycled PET, mild steel and Sn plates were used as substrates. Recycled PET plate was made from crushed PET bottles. The crushed PET bottles were then remelted and reformed into PET plate (5×5×4mm).

2.2 Experimental procedures

The coatings on the PET plate substrates were deposited by atmospheric plasma spraying with Cu and TiO_2 powders. The plasma spray conditions used for this research are shown in **Table 1** for Cu powders and **Table 2** for TiO_2 powders. The spraying distance and traverse speed of plasma spraying gun were changed to investigate the effect of spray conditions on the structure of deposited Cu and TiO_2 coatings and the implantation phenomena of Cu and TiO_2 particles into PET substrate. Cross sections of deposited layers and implantation of sprayed particles and surface morphology were observed by optical microscope and scanning electron microscope (SEM). The element distribution of implanted particles was examined by energy dispersive X-ray spectroscopy (EDX).

Table 1 Plasma spraying conditions for Cu

Spraying atmosphere	Air
Arc current	500 (A)
Arc voltage	55 (V)
Spraying distance	150~250 (mm)
Step width	6 (mm)
Traverse speed of gun	5.5 or 10 (m/min)

Table 2 Plasma spraying conditions for TiO_2

Spraying atmosphere	Air
Arc current	500 (A)
Arc voltage	60 (V)
Spraying distance	100, 200 (mm)
Step width	5 (mm)
Traverse speed of gun	9.2×10^{-2} (m/s)

3. Results and discussion

3.1 Effect of substrate property on the deposition procedure of sprayed Cu particles

Cu powders of 45~90μm sizes were sprayed on a mild steel substrate by atmospheric plasma spraying for a spray distance of 200mm and at the traverse speed of gun of 1.7×10^{-1}(m/s). **Figure 2** shows a typical optical microstructure of a cross section of plasma as-sprayed Cu coating on the mild steel. As expected, Cu coatings on mild steel showed a layer structure formed by the deposition of rapidly solidified flattened particles. In order to modify the surface property of the substrate made from waste PET bottles, plasma spraying was tried at the same condition used for mild steel. **Figure 3** shows the microstructure of a cross section for PET substrate with plasma sprayed Cu particles. From this figure, some Cu particles exist on the PET substrate surface. In spite of the lower melting point of PET plate, the heat damage caused for polyvinyl chloride plate was not seen for recycled PET plate after plasma spraying.

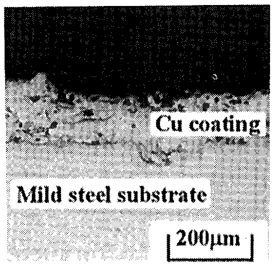


Fig.2 Microstructure of cross section of mild steel substrate after plasma spraying of Cu (Traverse speed: 1.7×10^{-1} m/s)

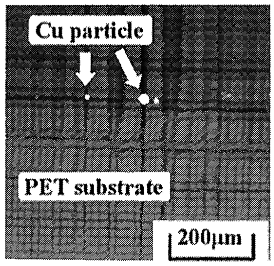


Fig.3 Microstructure of cross section of PET substrate after plasma spraying of Cu (Traverse speed: 1.7×10^{-1} m/s)

In order to increase the heat input for Cu particles and PET substrate, the traverse speed was reduced from 1.7×10^{-1}(m/s) to 9.2×10^{-2}(m/s). **Figure 4** shows the cross section of test pieces after plasma spraying of Cu powders under such spray conditions.

In this case a large amount of spherical Cu particles, without deformation, penetrated deeply into the PET plate surface in spite of melted liquid Cu particles. **Figure 5** shows the EDX analysis of the Cu element for cross sections of Cu sprayed test pieces of PET plate substrate after plasma spraying. From this figure, it is revealed in more detail that Cu particles existed in the PET substrate with spherical shape.

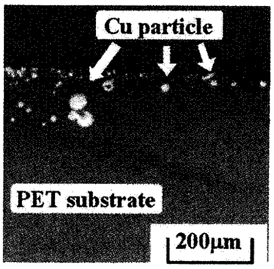


Fig.4 Microstructure of cross section of PET substrate after plasma spraying of Cu (Traverse speed: 9.2×10^{-2} m/s)

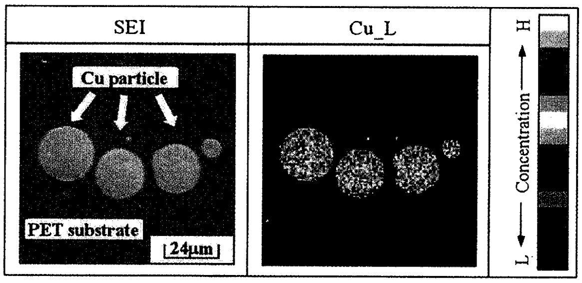


Fig.5 EDX map analysis results of cross section of PET substrate after plasma spraying of Cu

To make clear the difference of plasma spray phenomena between the mild steel and PET plate, Sn plate substrate with a melting point (230℃) lower than the PET substrate was selected for plasma spraying of Cu powders. **Figure 6** shows the microstructure of the cross section for a Sn substrate with plasma sprayed Cu coating. From this figure, it is revealed that the ordinary layered structure coating was formed on Sn plate substrate as well as the mild steel substrate. It was considered that the thermal spray phenomena were caused by the difference in the thermal conductivity of the substrate, rather than melting point and difference of conductivity, cause different surface states between PET and Sn substrates. In order to observe the surfaces of PET and Sn substrates with heat input, a plasma jet was used to irradiate both substrates. **Figure 7** shows the surface morphology of a PET substrate after plasma jet irradiation at the spray distance of 200mm without Cu powders. From this figure, it was found that the surface of PET substrate was melted and deformed during plasma jet irradiation. **Figure 8** shows the surface morphology of a Sn substrate 60after plasma jet irradiation at the spray distance of 200mm without Cu powders. From this figure, it was found that the surface of the Sn substrate was not deformed by melting during plasma jet irradiation. It was also observed by a high speed video camera that the surfaces of both substrates were melted during plasma spraying. However the surface of a Sn substrate could not keep a liquid state because of the higher thermal conductivity of the substrate, while PET plate was kept in the liquid state because of the lower thermal conductivity. So, liquid Cu particles could be implanted into the melted PET substrate surface without deformation.

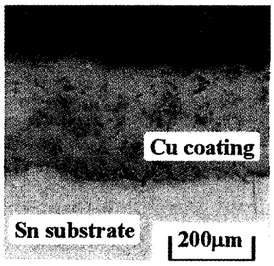


Fig.6 Microstructure of cross section of Sn substrate after plasma spraying of Cu (Traverse speed: 9.2×10^{-2} m/s)

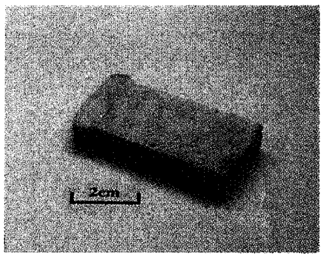


Fig.7 Surface morphology of PET substrate after plasma jet irradiation (Irradiation distance: 200mm)

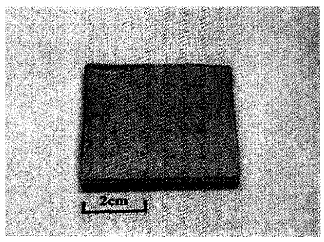


Fig.8 Surface morphology of Sn substrate after plasma jet irradiation (Irradiation distance : 200mm)

3.2 Effect of spray distance on the deposition procedure of sprayed Cu particles

In order to change the heat input of plasma spraying for PET plate substrates, spray distances were increased from 200mm to 300mm at a constant traverse speed of 9.2×10^{-2}(m/s). In the case of an irradiation distance 200mm, surface morphology of PET substrate was shown in Fig.7 from which it was found that the surface was melted and deformed during plasma jet irradiation. **Figure 9** shows surface morphology of a PET substrate after plasma jet irradiation at the spray distance of 250mm without Cu powders. It was found that melted zones on the PET substrate surface were not observed. **Figure 10** shows the microstructure of a cross section of a PET substrate after plasma spraying at the spray distance of 200mm with Cu powders. **Fig.11** shows a similar microstructure for the spray distance of 250mm with Cu powders. As shown in Fig.11, when the surface of the PET substrate was not deformed by melting during plasma irradiation with a low heat input, some of the Cu particles were observed inside the substrate However, in the case of a lower heat input at the spray distance of 300 mm, it was found that a Cu coating was formed as shown in **Fig. 12**.

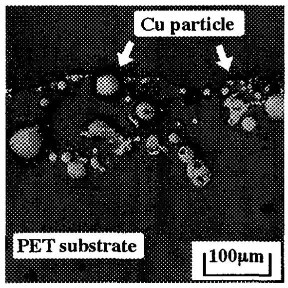


Fig.11 Microstructure of cross section of PET substrate after plasma spraying of Cu (Spray distance : 250mm)

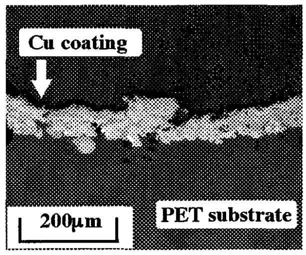


Fig.12 Microstructure of cross section of PET substrate after plasma spraying of Cu (Spray distance : 300mm)

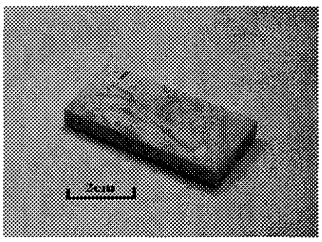


Fig.9 Surface morphology of PET substrate after plasma jet irradiation (Irradiation distance : 250mm)

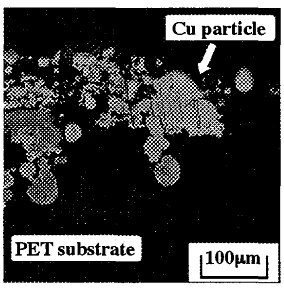


Fig.10 Microstructure of cross section of PET substrate after plasma spraying of Cu (Spray distance : 200mm)

In the case of the highest heat input at the spray distance of 200 mm, the schematic illustration of the deposition process of plasma spraying with Cu powders was shown in **Fig. 13**. As shown previously in Fig. 10, when the surface of the PET was melted completely during plasma spraying with a high heat input, a large amount of Cu particles penetrated deep inside the PET substrate. In the case of low heat input at the spray distance of 250 mm, the process of deposition procedure of sprayed Cu particles is shown in **Fig. 14**. In this case, in spite of no melting of PET substrate surface as shown in Fig. 9, some Cu particles were implanted just inside the PET substrate. As shown in Fig. 14, it was considered that the PET substrate surface was melted slightly due to the interaction of heat of plasma jet and melted flying Cu particles during plasma spraying and Cu particles were implanted into the PET substrate surface. The existence of Cu particles just inside the PET substrate occurred without deformation of the melted Cu particles. The plasma spraying of lower heat input at the spray distance of 300 mm results in the formation of a Cu coating. At the spray distance of 300 mm, melted Cu particles were deposited on the solid PET substrate surface without melting of the PET surface to produce a Cu coating.

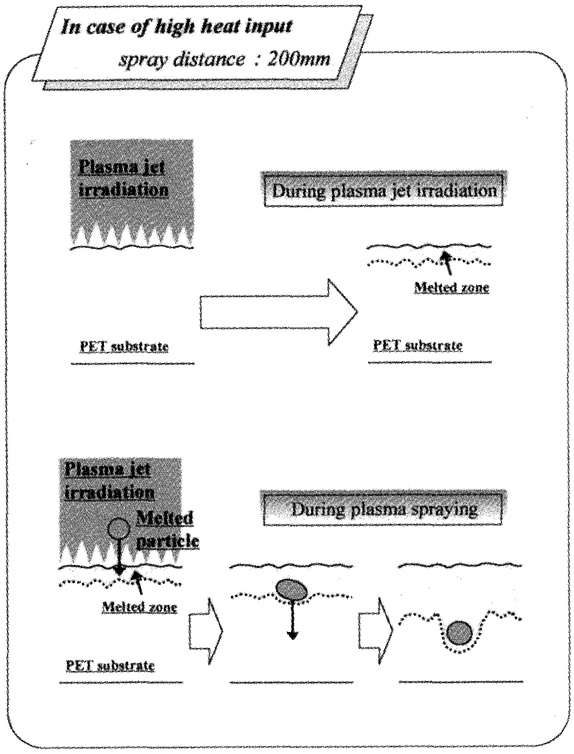


Fig.13 The deposition procedure of sprayed Cu particles at high heat input

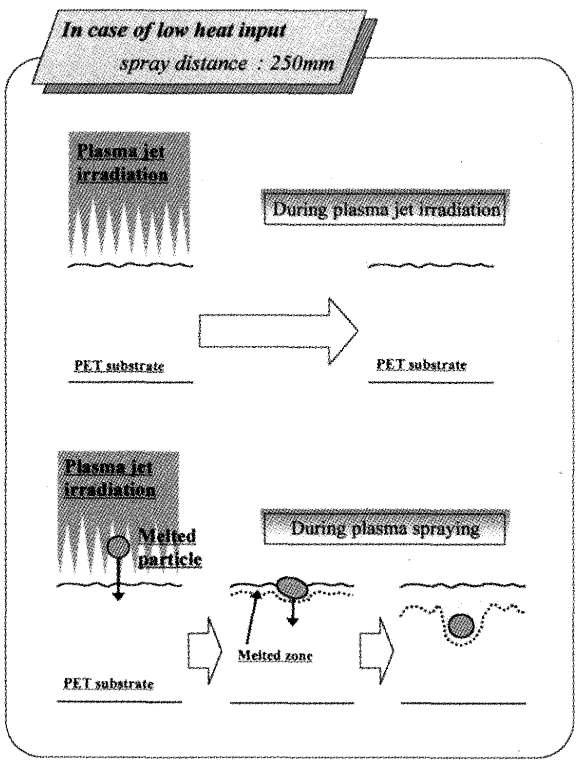


Fig.14 The deposition procedure of sprayed Cu particles at low heat input

3.3 Plasma spraying of TiO_2 onto recycled PET substrate

Rutile type TiO_2 was plasma sprayed onto a PET substrate under the condition of spray distance of 100 mm, which is often used in TiO_2 spraying. In this case, deformation and heat damage of the PET occurred as shown in **Fig. 15**. Then the spray distance was increased from 100 mm to 200 mm in order to decrease the heat input into PET substrate and TiO_2 particles. As a result, some spherical particles were implanted into the PET substrate as shown in **Fig. 16**. This figure also indicates that many flat particles accumulate on the implanted particles.

On the other hand, ordinary layered coatings were formed on mild steel and Sn substrate under the same spray conditions, as is shown in **Fig. 17 (a), (b)** respectively. Therefore, it was considered that the similar process as mentioned in 3.2 section was also operating during plasma spraying with TiO_2 powder.

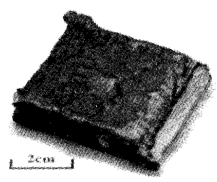


Fig.15 Surface morphology of PET substrate after plasma spraying of TiO_2

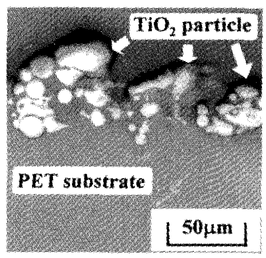
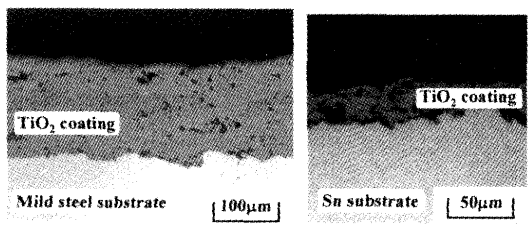


Fig.16 Microstructure of cross section of PET substrate after plasma spraying of TiO_2



(a) (b)

Fig.17 Microstructure of cross section after plasma spraying of TiO_2
(a) Mild steel substrate (b) Sn substrate

4. Conclusions

Recycling methods for waste PET bottles, that is, reproduction of PET plate and surface modifications, were developed using a plasma spray technique with Cu and TiO_2 powders. The effects of spraying conditions on the formation of functional coatings were also investigated. The results are summarized as follows.

(1) Plasma spraying onto recycled PET substrates with Cu and TiO_2 powders were carried out under various spraying conditions. As a result, particle implantation phenomena were observed under the conditions of high heat input. And ordinary layered coatings were formed as the heat input was decreased. Therefore, it is suggested that the structure of the coating can be controlled by the selection of spraying condition.

(2) The processes of particles implantation phenomena were examined. As a result, it was found that the dominant factor for coating phenomena during plasma spraying was the thermal conductivity of substrate instead of its melting point.

(3) Improvement of bond strength between substrate and coatings will be expected by accumulating various materials with wedge-like implanted particles.

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