



Article

# Coating Qualities Deposited Using Three Different Thermal Spray Technologies in Relation with Temperatures and Velocities of Spray Droplets

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Abstract: Three guns based on different thermal spray technologies—namely, gas flame spray, wire arc spray, and wire plasma spray—were operated at each best cost—performance condition, and the resulting spray droplets and deposited coating qualities were investigated. For the former, a simple optical monitoring system was used to measure temperatures and velocities of spray droplets ejected from the guns. On the other hand, for the latter, qualities of coating layers on substrates—namely, surface roughness, atomic composition, hardness, adhesive strength, and porosity—were characterized. Then, these coating qualities were discussed with respect to the measured temperatures and velocities of spray droplets, which revealed novel features in the coatings that have not been seen before, such as atomic composition and hardness strongly dependent on temperature and environments of droplets towards the substrates, and porosity on velocity of droplets impinging onto the substrates.

**Keywords:** spray droplet from wire; temperature; velocity; gas flame spray; wire arc spray; wire plasma spray; coating quality

## 1. Introduction

Thermal spray technologies have been used for the last several decades for various applications, such as for anti-corrosion of electricity transmission towers and energy generation, etc. [1], together with bridges [2], subsea rises/pipelines [3], and anti-wear for various contacting components [4]. As was pointed out by Fauchais and his coauthors [5], company workers of plasma spray have been trying to find good coating services (corrosion and wear resistance, thermal protection, etc.) and coating repeatability, reproducibility, and reliability for applications in the fields. This endeavor has to be carried out at the best cost–performance; that is, acceptable coating qualities for various purposes have to be pursued at reasonably competitive costs compared with other competitors/methods/techniques. On the other hand, researchers in laboratories tend to seek to understand coating processes and resulting coating microstructure and properties [5].

This article is a result of the joint effort of field workers in a thermal spray company who have been routinely employing thermal spray for more than a decade at the above-mentioned "best cost-performance", with university researchers who have been trying to narrow the existing gap of their effort with industry by supplying the most pertinent data for understanding and optimizing thermal spray processes. In the process of trying to find operating conditions of various thermal spray

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guns at the best cost–performance in the fields, wire arc spray has been observed the to yield, for example, fast deposition at relatively porous and soft coating layers, while gas flame spray to yield hard coating layers of low porosity at relatively low adhesive strength. On the other hand, coauthors of the present article at the University have believed that temperature and velocity of spray droplets to have profound effects on these characteristics/qualities of coating layers, and have spent much effort to realize a simple monitoring system for the purpose [6].

In this background, a joint work has been started to investigate the relationship of the qualities of coating layers deposited using three spray guns based on the different thermal spray technologies operated at their best cost–performances, in order to closely look at them with respect to temperatures and velocities ejected from the guns and impinging onto substrates. As the measures of characteristics/qualities of thus-deposited coating layers, the measured surface roughness, atomic composition, hardness, porosity, and adhesive strength were compared on the same comparative basis for the coating layers deposited over such wide ranges of operating parameters. As far as the present authors are aware, there has not been any work of this kind carried out and reported on in open literature in the past. Therefore, it is their belief that the results revealed novel features, such as composition and hardness heavily dependent on the temperature and production environment, and porosity on the velocity of droplets impinging on substrates. These results, together with other details, are described.

This article is structured as follows: Section 2 describes the feed stock and deposited material, methods, and equipment; Section 3 presents the measured results of droplet temperatures and velocities together with those of coating qualities; and Section 4 draws observations among these two values and the resultant discussions. Section 5 summarizes the results.

## 2. Material, Methods, and Equipment

## 2.1. Material and the Method of Surface Treatment for Thermal Spray Coating

Substrates on which coating layers were deposited were JISG3101 SS400 ( $75 \times 150 \times 6 \text{ mm}^3$ ), which were first blast-treated following the procedures described in ISO8501-1 Sa3.0 (surface roughness to be more than Ra 8.0  $\mu$ m and Rz 50.0  $\mu$ m). The substrates were blasted with F-24 Al<sub>2</sub>O<sub>3</sub> grit (NANIWA ABRASIVE MFG. Co., Ltd., Osaka, Japan) at a pressure of 0.6 MPa using a nozzle diameter of 6 mm at a standoff distance of 100 mm and an angle of attack of 90°. The type A-3R (ATSUCHI TEKKO Co., Ltd., Osaka, Japan) was used as the blasting equipment.

## 2.2. Productions of Spray Droplets Using Al/5Mg Wire and Spray Parameters for Coating Productions

As the material for the droplet formation of thermal spray, the present work employed an aluminum-5% magnesium alloy A5056 (hereafter referred to as Al/5Mg). This is widely used for anti-corrosion of iron and other surfaces. Table 1 shows the Al/5Mg wire composition. The gas flame gun used the wire with a diameter of 3.2 mm, while wire arc and wire plasma guns used that of 1.6 mm (two of this for the wire arc gun).

Material	Wire Diameter (mm)	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al
Al/5Mg	3.2	0.09	0.19	0.01	0.09	4.53	0.07	0.00	0.04	Balance
wire	1.6	0.13	0.17	0.00	0.05	4.83	0.06	0.00	_	Balance

**Table 1.** Composition of the coating material (wt.%).

As the methods of wire thermal spray, the three most widely used types of guns were employed in the present study—namely, gas flame, wire arc, and wire plasma. Figure 1 shows a cross-section of the "plazwire" gun head, which is a type of wire plasma system, and has been most extensively used in fields of the present authors' company [7]. The other two guns are of the conventional types—namely, a gas flame [8] and a wire arc [9]—and are not described any further here.

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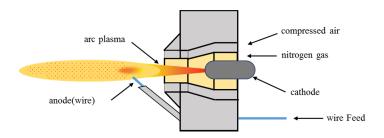


Figure 1. A cross-section of the "plazwire" system.

Table 2 gives the spray parameters of the three guns used for the coating productions.

Method	Equipment	Standard Spray Distance (mm)	Wire Feed Rate (kg/h)
Gas flame	Metco14E	150	4.0
Wire arc	Metallisation 150/s450	150	3.2
Wire plasma	PW-120	150	3.0

**Table 2.** Spray parameters used for coating productions.

2.3. The Methods and Arrangements of Equipment and Instruments for Measuring Temperature and Velocity of Spray Droplets

The method of measuring the temperature of spray droplets is well-known as a "two-color" thermometry, in which an intensity ratio at two wavelengths of an optical emitter is compared. For this purpose, the Planck emission formula [10] is used, which is written as follows:

$$I(\lambda, T) = \varepsilon(\lambda, T) \frac{2\pi hc^2}{\lambda^5} \frac{1}{\exp\left(\frac{hc}{\lambda kT}\right) - 1}$$
 (1)

Here,  $I(\lambda,T)$  is an intensity of radiation emitted at a wavelength of  $\lambda$  from a body surface at a temperature of T;  $\varepsilon(\lambda,T)$  is an emissivity which is dependent on the material and its surface conditions (such as surface roughness and the degree of oxidation), together with wavelength and the temperature of the emitting surface; h is the Planck's constant; c is the speed of light; and k is the Boltzmann constant.

This method is usually applied at visible and near-infrared regions of electromagnetic wave radiations. In order to use this method in practice, one has to know values of emissivity of the emitter surfaces at hand at the two wavelengths by referring to suitable references [11]. In order to implement the method, the emitted radiation from the surface is collected through two narrow bandpass filters, which are electronically measured. Thus, it is only a matter of calibrating the two measured intensities against temperature by correlating them using the Planck's formula and the surface emissivity.

The method of velocity measurement is also well known as a "time-of-flight (TOF)" technique, in which a time difference  $\Delta t$  of a passage of a radiating body between well-defined positions having a distance of  $\Delta x$  is measured to yield the velocity  $v = \xi \Delta x/\Delta t$ , where  $\xi$  is a magnification or reduction ratio of a collection optics of a radiating body onto a recording surface. Because the passage of an image of a radiating body on the recording surface is very brief, it has to be confirmed whether the time response of each pixel element of the recording surface is fast enough to catch a real-time history, which necessitates a calibration procedure.

Figure 2 shows the arrangement of the simple monitoring system, consisting of a sensor unit (a), a control board unit (b), and a PC for data analysis (c). The sensor head consisted of a light collecting lens followed by a part for separating the light into two wavelengths, with the light of each wavelength guided through an interference filter (central wavelengths of 650 nm and 750 nm with a bandwidth of 50 nm each) into a fiber-coupled silicon photo-diode (Si-PD) to be electronically detected.

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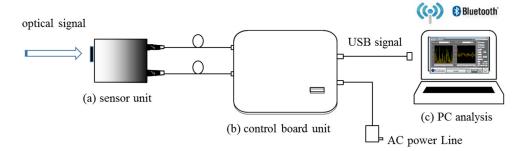


Figure 2. Arrangement of the simple monitoring system.

For the present purposes of obtaining relationships between measured temperture and velocity of spray droplets with coating qualities, discussions are concentrated on the results at the measurement position of temperature and velocity at the central position of substrates for the deposition of coating layers, namely 150 mm downstream and on the center-line of the spray guns.

# 2.4. Methods of Coating Depositions and Employed Techniques of Measuring Coating Qualities

Coating layers were deposited using three thermal spray technologies of combustion gas flame, wire arc, and wire plasma spraying, using Al/5Mg wires shown in Table 1 to coating thicknesses of approximately  $100~\mu m$ .

The methods of measuring coating qualities are as described below, with their validations mostly approved by the JIS (Japanese Industrial Standards) and the ISO (International Organization for Standardization), as shown below with respective numbers.

First, the surface roughness was measured using Surftest SJ-201 (Mitutoyo Corporation, Kanagawa, Japan) following the JISB0601 "2001" instruction.

Atomic composition was observed using a scanning electron microscope (SEM) instrument for the cross-sectional view using 15.0 kV  $\times$  100 times magnification, and an electron probe micro analyzer (EPMA) instrument (JXA-8500F, JEOL Ltd., Tokyo, Japan) for atomic concentration with their semi-quantitative analysis. During the analyses and measurements, "the instructions for users" were rigorously followed.

Hardness was measured using a Micro-hardness tester (MVK-G1, Mitutoyo Corporation, Kanagawa, Japan) at a load of 100 g for sample numbers of n = 10. During the analyses and measurements, "the instructions for users" were rigorously followed.

Adhesive strength was measured following the JISH8300 instructions (in accordance with ISO2063) using Techno-tester2000LD (SANKO-TECHNO Co., Ltd., Chiba, Japan).

Porosity was measured following the JISR1643 instructions, where an open-pore measurement technique was employed, namely by measuring weight changes using distilled water in vacuum, among sample numbers of n = 3.

# 3. Results of the Measurements

# 3.1. Temperature and Velocity of Spray Droplets

Table 3 summarizes the measured values of temperatures and velocities. It is to be noted that there is an uncertainty of up to 20% overestimate in these temperatures [6] and 10% underestimate in these velocities. The overestimates come from uncertainty in the emissivity value [11] of droplets, and the underestimates come from the necessity of subtracting the contributions of low temperature droplets existing at the peripheries of the droplet flow which are inevitably observed for this kind of line-of-sight measurements (this subtraction was not carried out in the present values).

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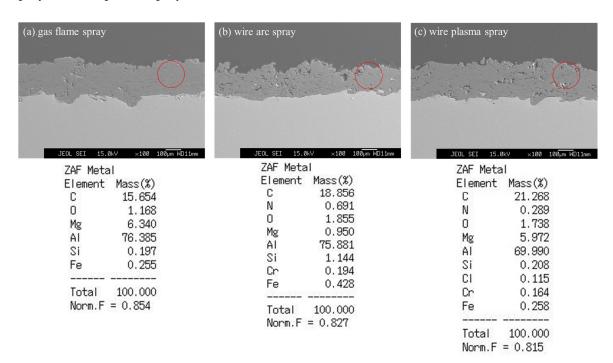
**Table 3.** Measured temperatures and velocities of spray droplets ejecting from three different thermal spray guns.

Method	Temperature (K)	Velocity (m/s)
Gas flame	1900	200
Wire arc	2900	60
Wire plasma	3000	90

It is to be noted that these measured values are broadly in agreement with those obtained using commercially available instruments [12,13]. However, the results shown in Table 3 were obtained after careful calibration of the present instrument [6] with confirmed accuracies with two significant digits. In addition, the above uncertainties in temperature and velocity were only available after obtaining raw data by the present authors.

# 3.2. Qualities of Coating Layers

The three photographs in Figure 3 show the cross-sectional views as observed using the SEM method. These results do not show any marked difference for coating layers deposited using the three thermal spray guns. The red circles in the figures show the positions of semi-quantitative analyses for the EPMA method. From the atomic compositions, one recognizes an almost complete disappearance of Mg for the wire arc spray case, while the Mg compositions were almost the same for both gas flame spray and wire plasma spray cases.



**Figure 3.** Cross-sectional views obtained using SEM (above) and the results of the semi-quantitative analyses (following the instruction commonly referred to as the "ZAF" corrections) using an electron probe micro analyzer (EPMA) for coating layers deposited using the three thermal spray guns (below, which are direct-printouts pasted here from the EPMA instrument). (a) Gas flame spray; (b) Arc spray; (c) Plasma spray.

Table 4 shows the values of the measured surface roughnesses, hardnesses, adhesive strengths and porosities obtained for coating layers deposited using the three thermal spray guns. The surface roughnesses were  $R_z$  80–100  $\mu$ m for both combustion gas flame and wire plasma cases, were a little

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smoother for the former (gas flame case), and  $R_z$  150  $\mu$ m for the wire arc case. The mesured average hadnesses were, in the harder order, HV = 69.5 for wire plasma spray, 65.3 for gas flame spray, and 41.4 for wire arc spray. Measured average adhesive strengths were, in the stronger order, 14 MPa for wire arc spray, 7.5 MPa for wire plasma spray, and 6.6 MPa for gas flame spray. Finally, the measured porosities were, in the more porous order, 24% for wire arc spray, 19% for wire plasma spray, and 14% for gas flame spray.

**Table 4.** Results of the measured roughnesses, hardnesses, adhesive strengths, and porosities obtained for coating layers deposited using the three thermal spray guns. In this table, "-" indicates the upper and lower values of two measurements, and " $\pm$ " indicates the ranges of more than three measurements with their average values at each front.

Method	Surface Roughness R <sub>z</sub> (μm)	Hardness (HV)	Adhesive Strength (MPa)	Porosity (%)
Gas flame	80–100	$65.3 \pm 15.0$	6.61-6.65	$14 \pm 1.0$
Wire arc	130-150	$41.1 \pm 8.0$	13.4–14.5	$24 \pm 2.0$
Wire plasma	80–100	$69.5 \pm 14.0$	7.50–7.85	$19 \pm 2.0$

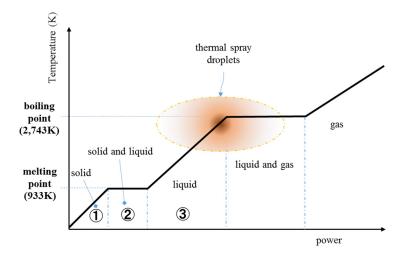
#### 4. Discussions

# 4.1. Power Balances in the Three Thermal Spray Guns

It is of interest to know how much power is dissipated in a thermal spray gun, and how much of it is used to heat spray droplets impinging onto substrates. For this purpose, measured values of temperatures were used together with known input powers. The heat loss from a gun head to the surrounding air was calculated to be negligible from the measured surface temperatures of the gun head and the surrounding ambient air.

The powers to gun heads are obtainable from electrical input powers for plasma- and arc-heated guns, while from a gas flow-rate and a heat of combustion for a gas-heated gun where complete burning of the supply gas is assumed.

On the other hand, the powers to spray droplets are obtainable by referring to Figure 4; from feed-rates of wires multiplied by (1), a sensible heat to raise temperatures of wires from an ambient temperature to the melting point of 933 K for Al (neglecting the contribution of 5% Mg), (2) a latent heat at the melting point (397 kJ/kg), and (3) a sensible heat to raise temperatures of wires from the melting point to measured values. The power efficiencies are obtainable from  $\eta = ((1) + (2) + (3))/(input power)$ .



**Figure 4.** Schematic diagram showing how an input power to a gun is used to heat up spray droplets (abscissa is in a relative unit). The melting and boiling points in the ordinate is for Al.

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The obtained results are shown in Table 5. As seen from this table, efficiencies are, in the order of higher efficiency, 45% for arc-heated, 23% for plasma-heated, and 9% for gas-heated guns. The implications of this result are discussed in the latter half of Section 4.3.

**Table 5.** Powers to guns and various heating powers to spray droplets, together with power efficiencies for three thermal spray guns.

Method	Input Power (kW) _	Input Power to Thermal Spray Droplets (kW)			Total Power into Spray Droplets (kW)	Efficiency (%)
		Step (1)	Step (2)	Step (3)		(70)
Gas flame	18.0	0.51	0.35	0.75	1.61	9
Wire arc	7.3	0.71	0.44	2.13	3.28	45
Wire plasma	10.0	0.48	0.33	1.53	2.34	23

# 4.2. Coating Qualities in Relation with Temperatures and Velocities of Spray Droplets

# 4.2.1. Gas Flame Spray Gun

Gas flame sprayed droplets are characterized by a low temperature (about 1900 K) and a high velocity (about 200 m/s), as seen in Table 3. The first evidence observed in Figure 3 is that the atomic composition of coating layers has hardly changed from that of the supplied wire, which is believed to be due to almost no loss of Mg due to the low temperature of droplets. The high hardness of coating layers as shown in Table 4 is explained by this maintenance of the Mg composition. The low adhesive strength may have resulted from the low temperature of spray droplets. The low porosity of the coating layers in the same table is also explained by the high velocity of spray droplets impinging onto a substrate.

# 4.2.2. Wire Arc Spray Gun

Wire arc sprayed droplets are characterized by a high temperature (about 2900 K) and a low velocity (about 60 m/s), as seen in Table 3. As evidently observed in Figure 3, the Mg in the coating layers has almost disappeared due to its low boiling temperature of 1364 K, and a small amount of nitrogen has appeared. The very low hardness of coating layers—almost similar to Al, as shown in Table 4—is believed to be due to the almost complete loss of Mg by the high temperature of droplets. The high adhesive strength may be due to the high temperature of the spray droplets. The high porosity of the coating layers may have resulted from the low velocity of spray droplets impinging onto the substrate.

## 4.2.3. Wire Plasma Spray Gun

Wire plasma sprayed droplets are characterized by a high temperature (about 3000 K) and an intermediate velocity (about 90 m/s), as seen in Table 3. The atomic composition of coating layers as shown in Table 4 has not changed much from that of the supplied wire in spite of the high temperature of spray droplets, similar to arc-heated droplets. This evidence, together with the high hardness of coating layers (e.g., Table 4), may have resulted from the presence of a shielding gas supplied around and along the flow of the spray droplets, which is intended to suppress mixing of spray droplets with the surrounding ambient air. The observed fact of lower adhesive strength of coating layers in the same table, compared with those of arc-heated droplets—despite the similar high temperatures for both cases—may have resulted from other unidentified cause(s). The porosity of the plasma-heated case (also from the same table) is between the other two cases, which may be due to the intermediate values of the temperature and velocity.

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## 4.3. Overall Assessment of the Obtained Results

There have been extensive studies and resulting literatures for obtaining good coating layers by thermal spray processes (a good overview of a history up to the present time is given in [5], together with what have been carried out from the standpoint of monitoring thermal spray processes) and for practical applications in various fields [1–4]. These studies are mostly concerned with particular types of guns for understanding thermal spray processes with its optimization and/or for specific applications. This article is distinctly different from these past studies in that three guns of completely different technologies were employed. Their dimensions and operating conditions were optimized, as described in Section 1, for "the best cost–performance" after much trial-and-error, and are now routinely in use for depositions of corrosion- and wear-resistant layers in field works of the present coauthors' company. The wide ranges of temperatures and velocities as shown in Table 3 together with wide ranges of coating qualities as evident from Table 4 and of power efficiency as shown in Table 5 are only possible through the present approach. It is to be noted that the values of coating qualities shown in Table 4 may not be as good as one may hope, but it is the result of "the best cost–performance" choice, as one may obtain better qualities at the expenses of other factors, such as cost and/or coating time.

The observed results of coating qualities in relation with temperature and velocity of spray droplets were described above in Section 4.2, and a few words may be of value on the power efficiencies shown in Table 5. It is to be noted from Table 5 that the power efficiency of the wire arc spray gun is the highest at 45% among the three guns, followed by the wire plasma spray gun at 23%, then by the gas flame spray gun at 9%. These results are well understandable, as follows: power to the wire arc spray is supplied directly outside the gun, unhindered much from the compressed air sent to the arc region to transfer spray droplets to substrates, with the resultant power to the wire approaching almost half of the supplied amount. On the other hand, power to the wire plasma spray is first used to ionize the supplied nitrogen gas, and thus-formed plasma reaches the anode, which is in fact the wire to become spray droplets after melting. In this process, almost three-quarters of the supplied power is taken away by the ionized gas (plasma), and about one quarter remains in the droplets. Finally, power to the gas flame spray is only through heat transfer from burnt gas to the wire, which is very slow and of indirect process compared with electric discharges towards the wire electrodes, resulting in only less than 10% of the supplied power transferred to formed droplets for this case.

These features yield the following obvious outcomes: fast production of spray droplets and coating layers deposited by the wire arc spray, with rather rough surface and high porosity due to low velocity of the droplets resulting from their poor acceleration by the compressed air. High velocity of spray droplets in gas flame spray is the result of their good acceleration in a well-constricted nozzle, where the constricted nozzle shape is necessary to make as much thermal contact of burnt gas with the central wire. This high velocity yields low porosity of the coating layer. For the case of the wire plasma spray, the nozzle is constricted as shown in Figure 1 in order to yield good acceleration there, but not to the extent of the small annular structure for the gas flame spray. Additionally, a compressed air is supplied (as shown in Figure 1) to shield and prevent the outside air from mixing and decelerating the spray droplets' flow in the central core. The authors believe this feature to have played an important role in keeping the atomic composition as shown in Figure 3 and the resulting hardness as shown in Table 4.

The results of power efficiencies shown in Table 5 and the resulting spray qualities shown in Figure 3 and Table 4 are summarized as the wire plasma spray to be placed in between the gas flame spray and the arc wire spray in almost every aspect. Therefore, these characteristics are to be considered when considering which technology is to be selected for a particular application.

### 5. Conclusions

From the comparative studies of coating qualities deposited using three thermal spray guns with the measured temperatures and velocities of spray droplets impinging onto the substrates for the three Coatings 2017, 7, 27 9 of 10

cases, the following conclusions were obtained, summarized in terms of coating qualities, rather than from the viewpoints of various guns as described in Section 4.

- Atomic Composition. This is believed to be decided by the atmosphere when spray droplets are formed, and by their temperatures. Spray droplets produced by the wire arc spray-heating are formed in an open atmosphere and at a high temperature, resulting in a deposited atomic composition much altered from that of the supplied wire. On the other hand, discharges in the wire plasma spray-heating are maintained in nitrogen gas, and the flow of thus-formed droplets are surrounded by a shielding gas, with the result that there is little composition change from that of the wire. Spray droplets produced by the gas flame spray-heating are formed at a low temperature, such that the deposited atomic composition is also hardly changed from that of the supplied wire.
- Hardness. This is believed to be mostly dictated by the atomic compositions of formed coating layers. In particular, the coating layers deposited using Al/5Mg is decided by the Mg composition, as evident for coating layers using all three different thermal spray guns.
- Adhesive Strength. This is believed to be decided by the temperatures of spray droplets, resulting in high adhesive strengths at high temperatures. However, the reason for different adhesive strengths deposited using wire arc spray- and wire plasma spray-heating—despite their similar temperatures—is not known at the present time. A weak adhesive strength deposited using gas flame spray-heating is well understood by a low temperature of spray droplets.
- **Porosity.** This is believed to be mostly dictated by the velocities of spray droplets, resulting in low porosities at high velocities and vice versa.

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**Author Contributions:** Yasuyuki Kawaguchi conceived and designed the experiments, together with writing the paper, with an advice and support of Fumihiro Miyazaki; Masafumi Yamasaki performed the experiments; Yukihiko Yamagata and Nozomi Kobayashi also joined the experiments, together with analyzing the data; Katsunori Muraoka contributed the overall strategy of this work and wrote the paper.

**Conflicts of Interest:** The authors declare no conflict of interest.

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