Proceedings of Korean Nuclear Society Spring Meeting
Seoul, Korea, May 1998

INDUCTION PLASMA DEPOSITION TECHNOLOGY FOR NUCLEAR FUEL FABRICATION

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Abstract

A study on induction plasma deposition with ceramic materials, yttria-stabilized-zirconia ZrO_2-Y_2O_3 *(m.p. 2640 °C), was conducted with a view of developing a new method for nuclear fuel fabrication. Before making dense pellets of more than 96%T.D., the spraying condition was optimized through the process parameters, such as chamber pressure, plasma plate power, powder spraying distance, sheath gas composition, probe position, particle size and powders of different morphology. The results with a 5mm thick deposit on rectangular planar graphite substrates showed a 97.11% theoretical density when the sheath gas flow rate was* Ar/H_2 *120/20 l/min, probe position 8cm, particle size* $-75\mu m$ *and spraying distance 22cm by AMDRY146 powder. The degree of influence of the main effects on density were powder morphology, particle size, sheath gas composition, plate power and spraying distance, in that order. Among the two parameter interactions, the sheath gas composition and chamber pressure affects density greatly. By using the multi-pellets mold of wheel type, the pellet density did not exceed 94%T.D., owing to the spraying angle.*

1. Introduction

Korea is unique in her reactor strategy, having adopted both PWRs and CANDUs. The synergy between the fuel cycles of PWR and CANDU reactors provides Korea with the potential merit of increasing the overall utilization of uranium and reducing the overall volume of spent fuel. On this basis, a fuel cycle concept, named DUPIC(Direct Use of spent PWR fuel In CANDU reactors) have been in experimental study under international cooperation with Canada and the USA. The IAEA has also recently joined the program. The OREOX(Oxidation and Reduction of Oxide fuel) process was chosen to be the most promising option for DUPIC fuel fabrication, being a process which does not involve any separation of sensitive materials and fission products[1-4]. Because of the high radiation fields emitted from DUPIC fuel, all processes have to be performed in shielded facilities. It is therefore required that all fabrication processes be simple. In Korea, a lot of efforts are being devoted to the development of new technologies. Plasma spraying is a valuable technique for obtaining strong (3040 MN m^{-}) and dense (90-99%T.D.) films by spraying ceramic or metal powders on the surface of materials at high speed through a plasma jet[5]. Compared to other spraying methods, like flames, arcs and explosions, the plasma technique can generate ultra-high temperatures nearing 20000 K to serve as a heat source and is an amazing method to melt and spray high melting point materials such as tungsten, molybdenum and various ceramics. In this study, the possibility of forming pellets with cylindrical shapes and high densities through the induction plasma spraying technique was evaluated as an alternative to the conventional multi-cycle oxidation/reduction and sintering processes, which are to be bottleneck, for DUPIC fuel fabrication. As well, those parameters having an influence upon the deposition properties were investigated.

2. Experimental System

2.1 Powder Material

As a surrogate for actual UO_2 powder, yttria-stabilized-zirconia(YSZ), including 20% Y_2O_3, was used. Two different morphology powders, METCO202NS (agglomerated, METCO Westbury, NY, USA) and AMDRY146 (sintered and crushed, Alloy Metal Co., Michigan, USA), were compared to investigate the effects on deposit density.

2.2 Equipment of Induction Plasma Spraying

100kW induction plasma equipment equipped with a 300kHz r.f. plasma torch was used in this study. Ar gas was used as plasma gas, and H_2 or N_2 gas with Ar was used as sheath gas. Figure 1 is the spraying chamber with double walls and enforcing water cooling system.

2.3 Molds for Pellet Formation

Figure 2 shows various kinds of specimen molds used in these experiments. Type 'A' mold, which is a rectangular type graphite bar, 25mm x 25mm x 40mm length, is generally used for optimizing the experimental parameters before fabricating the pellets. The other molds were also made of graphite and prepared for making real-sized pellets with 1 hole, 6 or 7 holes, 18 holes and 108 holes. The hole is the same size as a nuclear pellet: diameter 10mm x 10mm depth.

3. Results and Discussions

3.1 Optimization of Spraying Conditions for METCO202NS by ANOVA Method

Table 1 shows the density variation of the deposits, sprayed YSZ202NS powder with combination of 4 parameters, chamber pressure(A), plate power(B), spraying distance(C) and the composition of the sheath gas(D) at the 2 levels.

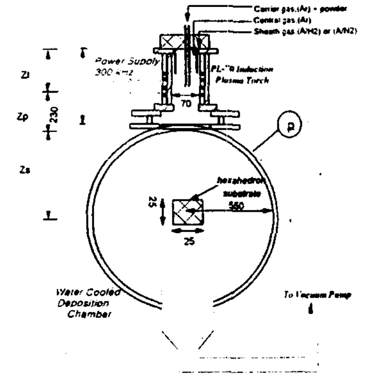

Fig 1. Schematic drawing of the powder deposition chamber

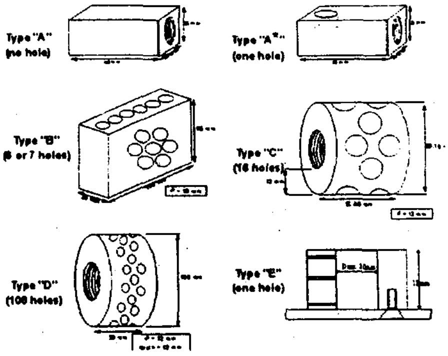

Fig 2. Schematic of the different sample substrates

Condition 'I' identifies the standard for these experiments, 200Torr, 80kW, 30cm, and Ar/H_2. Condition 'a' means the pressure parameter on 'I' changed to another level. Table 2 shows the results of the main and interaction effects and the F-value obtained from the ANOVA method from the experimental results of Table 1[6]. 'A' denotes the relative effect of variable A, 'B' denotes the effect of variable B, and 'AB' denotes the combined effects of variables A and B together. According to the results in Table 2, the sensitive parameters on the deposit density were sheath gas composition, plate power, spraying distance and chamber pressure, in that order. This indicates that the plasma temperature is the most important factor for high melting point ceramics. The thermodynamic properties of plasma such as plasma mass density, internal energy, enthalpy, specific heat and entropy, relate to the plasma gas composition[7]. In this experiment, the effects of the plasma gas composition to the deposit density were investigated using Ar/H_2 (120/10 l/min) and Ar/N_2 (100/40 l/min). The results by ANOVA indicate that the plasma gas composition is a dominant parameter for the deposit density, meaning that the gas composition is more influential than other factors due to the thermodynamic properties. The density of the deposit is revealed to be not so much dependent on chamber pressure as other main factors. However as shown in Table 1, the lower the chamber pressure was generally, the higher was the deposited density. Of the factor interactions, both the combination of the spraying distance and sheath gas composition, source 'CD', and chamber pressure and sheath gas composition, source 'AD' are very influential factors to the density of the deposit. For this experiment of plasma spraying with YSZ METCO202NS, the highest density of 90.08% T.D. was obtained at the following conditions: chamber pressure 200Torr, plate power 80kW, sheath gas composition Ar/H_2(120/10l/min) and powder spraying distance 22cm(the condition 'c' on Table 1).

Table 1. Design of the power deposition experiments of METCO202NS and its results

Effect	Treatment Combinations				Average
	A Pa (Torr)	B Pw (Kw)	C Zs (cm)	D Gx	Density (%)
I	200	80	30	Ar/H2	88.58
a	400	80	30	Ar/H2	83.69
b	200	60	30	Ar/H2	79.38
ab	400	60	30	Ar/H2	58.21
c	200	80	22	Ar/H2	90.08
ac	400	80	22	Ar/H2	88.29
bc	200	60	22	Ar/H2	86.17
abc	400	60	22	Ar/H2	80.65
d	200	80	30	Ar/N2	73.64
ad	400	80	30	Ar/N2	80.78
bd	200	60	30	Ar/N2	63.47
abd	400	60	30	Ar/N2	55.60
cd	200	80	22	Ar/N2	71.34
acd	400	80	22	Ar/N2	71.65
bcd	200	60	22	Ar/N2	51.17
abcd	400	60	22	Ar/N2	66.29

Table 2. ANOVA for power deposition experiments of METCO202NS

Source	Effect	Sum of Squares	Degree of freedom	Mean Squares	F value	Signi-fic ance
A	2.333	65.310	1	65.310	8.431	0.007
B	13.389	2151.103	1	2151.103	277.692	0.000
AB	-2.526	76.583	1	76.583	9.886	0.004
C	-2.785	93.102	1	93.102	12.019	0.002
AC	4.363	228.420	1	228.420	29.487	0.000
BC	4.120	203.734	1	203.734	26.301	0.000
ABC	-5.296	336.603	1	336.603	43.453	0.000
D	15.244	2751.997	1	2751.997	355.263	0.000
AD	6.009	433.261	1	433.261	55.931	0.000
BD	-1.832	40.278	1	40.278	5.200	0.029
ABD	-2.475	73.532	1	73.532	9.493	0.004
CD	-6.046	438.685	1	438.685	56.631	0.000
ACD	0.324	1.258	1	1.258	0.162	0.690
BCD	1.665	33.250	1	33.250	4.292	0.046
ABCD	2.160	55.966	1	55.966	7.225	0.011
ERROR		247.884	32	7.746		
TOTAL		7230.968				

3.2 Optimization of Spraying Conditions for AMDRY146 Powder

The effects of parameters such as H_2 flow rate in a sheath gas of Ar/H_2, probe position in the torch, particle size and spraying distance were investigated. As shown in Table 3, the highest density of 97.11%T.D. was obtained at the parameters; sheath gas of Ar/H_2 flow rate 120/20 l/min, probe position 8cm, particle size $-75\mu m$ and spraying distance 22cm. According to the ANOVA conducted with two kinds of particle sizes, $-75\mu m$ and $-90\mu m$, the deposit density is most dependent on the particle size, powder spraying distance, and H_2 flow rate in the sheath gas in this order, as shown in Table 4. In the case of two effect interactions, the combination of particle size and powder spraying distance, source 'CD', is the most influential to the deposit density.

3.3 Forming a Surrogate Pellet for Development of Nuclear Fuel

Based on the results of spraying METCO202NS powder and AMDRY146 powder on a fixed substrate, the pellet forming was conducted with pellet molds, as shown in Figure 2 and 3. The density of the pellet made in the revolving mold was 94% due to an unexpected factor and that was a little less than the previous one on the fixed substrate. The reason for the lower density was believed to be that the direction of the molten particle was not coincident to the longitudinal direction of the hole during spraying. However the shape of the pellet was excellent, as shown in Figure 4. Although real fuel materials were not used in these experiments, it is very meaningful to have applied a plasma technique to nuclear fuel fabrication. As shown

Table 3. Powder deposition experiments and results with -75 and -90μm AMDRY146

Effect	Treatment Combinations				Average Density (%)
	Sheath gas Ar/H2, (slpm)	Zp (cm)	particle size, m	Zs (cm)	
I	120/20	8	-75	30	95.51
a	120/10	8	-75	30	93.27
b	120/20	4	-75	30	94.49
ab	120/10	4	-75	30	89.76
c	120/20	8	-90	30	86.67
ac	120/10	8	-90	30	82.54
bc	120/20	4	-90	30	84.72
abc	120/10	4	-90	30	80.04
d	120/20	8	-75	22	97.11
ad	120/10	8	-75	22	94.64
bd	120/20	4	-75	22	96.60
abd	120/10	4	-75	22	94.25
cd	120/20	8	-90	22	94.80
acd	120/10	8	-90	22	89.15
bcd	120/20	4	-90	22	93.70
abcd	120/10	4	-90	22	87.50

Table 4. ANOVA for power deposition experiments of AMDRY146 -75 and -90μm

Source	Effect	Sum of Squares	Degree of freedom	Mean Squares	F value	Signi-fic ance
A	4.054	197.235	1	197.235	39.531	0.000
B	1.579	29.925	1	29.925	5.998	0.020
AB	-0.433	2.253	1	2.253	0.452	0.506
C	7.063	598.688	1	598.688	119.993	0.000
AC	-1.111	14.807	1	14.807	2.968	0.095
BC	-0.221	0.585	1	0.585	0.117	0.734
ABC	-0.158	0.301	1	0.301	0.060	0.808
D	-5.092	311.203	1	311.203	62.373	0.000
AD	-0.112	0.150	1	0.150	0.030	0.864
BD	0.667	5.333	1	5.333	1.069	0.309
ABD	-0.326	1.274	1	1.274	0.255	0.617
CD	2.702	87.642	1	87.642	17.566	0.000
ACD	0.648	5.044	1	5.044	1.011	0.322
BCD	0.242	0.701	1	0.701	0.140	0.710
ABCD	-0.326	1.274	1	1.274	0.255	0.617
ERROR		159.660	32	4.989		
TOTAL		1416.076				

in the previous results, it will be possible to form high density pellets of 96%T.D, as is required in nuclear fuel, by induction plasma spraying.

4. Conclusion

In the experiments to investigate the effects of various process conditions, such as chamber pressure, the plasma plate power, powder spraying distance and plasma gas composition to the density of thick deposit using METCO202NS powders, the plasma gas composition and plate power were found to be the most influential on the density. That is, the plasma temperature is an important factor for high melting point ceramics. In the case where two parameters interact, the combination of sheath gas composition and powder spraying distances greatly affect the density. In the optimization of AMDRY146 with various process parameters such as sheath gas composition, probe position, particle size and spraying distance, the highest density of 97.11 %T.D. was obtained at the following parameters: sheath gas flow rate 120/20 l/min, powder spraying probe position 8cm, particle size -75μm and spraying distance 22cm. Comparing this to the results of METCON202NS, the powder properties and particle size were found to be very influential on the density. At the optimum condition induced with a fixed substrate, pellets having a density of 94%T.D. with a nice exterior view, were formed.

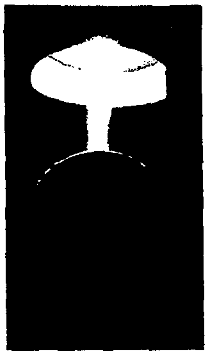

Fig 3. Induction plasma spraying of YSZ powder on Type "D" mold

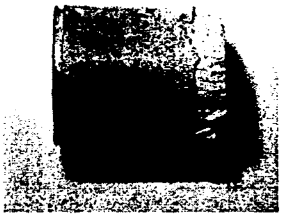

Fig 4. Pellet of YSZ of 10mm Dia. X 10mm H

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