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Thixoforming of 6066 aluminum alloy by multi-layer spray deposition①

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[Abstract] Two thixoforming technologies of 6066 aluminum alloy (Al-1.37Si-1.37Mg-0.77Cu-0.07Mn) produced by multi-layer spray deposition process were studied. The spray-formed materials are of equiaxed and very fine grain (10~20 μm). And the grain size coarsens slower than that of conventional casting materials at temperature below the liquidus, which may relate to high temperature particles distributed along the grain boundaries. Extrusion and hot pressing were used as the thixoforming processes respectively. After extrusion the materials show a microstructure of mean grain size below 20 μm without obvious recrystallization. The mechanical properties achieved via extrusion and pressing in semi-solid state attain that of common wrought materials with shorter peak aging time of 4~5 h, about half of that in conventional condition.

[Key words] thixoforming; multi-layer spray deposition; 6066 aluminum alloy
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1 INTRODUCTION

1.1 Thixoforming

In the early 1970s, a special thixotropic microstructure in semisolid state was discovered when Flemings and his co-workers studied the "hot tears" of the casting alloys[1]. The apparent viscosity of the semisolid materials declines sharply with the increase of the shear rate and time of mechanical agitation, which results in the microstructure of spheroidized particles distributed in the liquid without solid network. Through the strong agitation, the forming of solid network is greatly postponed, which gives enough time to conduct forming with very low stress. As-produced material can flow and fill the cavity of complex shape steadily at lower temperature in contrast with conventional casting, the technology in which components are produced in the semisolid state without solid network is termed "thixoforming", it brings the advantages as follows.

1) Better energy efficient because the process is easily automated and controlled, with production rates similar to or higher than pressure die-casting.

2) Smooth filling of the die with less air entrapment and low shrinkage porosity, gaining parts of high integrity and allowing the employment of heat treatable alloys possessing superior mechanical properties.

3) Less thermal shock to the die, resulting in longer die life and the possibility of semisolid forming of the high melting point alloys such as steels and superalloys.

4) Finer and more uniform microstructures, again leading to enhanced properties.

5) Higher yield resulting from the raw slug and mass savings in the components because of improved design, making the process cost effective.

The benefits mentioned above ensure that the thixoforming is a commercially viable manufacturing route. In fact, some components have been applied in the car industry on a commercial scale. As-produced parts are of reliable quality[2,3]. Manufacturers have found obvious cost savings as a result of high production rate and dramatically reducing subsequent machining operations.

1.2 Spray deposition

The spray deposition technology was firstly invented by Singer in the early 1970s, through which the materials with rapid solidification microstructure and big dimension can be produced directly. The mechanical properties improved due to their fine equiaxed grain and large excess solution degrees. This process had the advantage over the rapid solidification powder metallurgy technology in production of large parts with high properties. But only parts of simple shape can be produced up to now[4]. Furthermore, a small quantity of pores was difficult to eliminate. So subsequent processing such as hot extrusion and hot rolling and machining is necessary. The hot plastic deformation needs large scale press equipment in accordance with the good mechanical properties and thus leads to cost inflation. There are only several applications in military and expensive components.

A novel route combining spray deposition with thixoforming was recently studied[5~7]. It is feasible

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because the microstructures of spray deposition materials are characteristic of fine equiaxed grain in both fully solid and partially solid state (as will be discussed later). This combination may bring the advantages as: 1) cost savings for preparing thixotropic billets with high production rate and avoidance of consuming agitating equipment; 2) excellent microstructure with finer grain and very low coarsening rate in the state of semisolid, which gives as-processed materials better performance; 3) a flexible process amenable to producing metal matrix composites and new type of alloys with larger solution content than equilibrium solvability such as hypereutectic Al/Si alloys[6,7].

Preliminary research[5] has shown promising results. This paper focus attention on the phenomena about the microstructure and mechanical properties of 6066 aluminum alloy by the new route.

2 EXPERIMENTAL

2.1 Spray deposition

Multi-layer spray deposition technology[8,9] was adopted in this project. It differs from the traditional spray deposition technology by that the atomization cone moves repeatedly with its axis perpendicular or fixed-angle to substrate. Both the atomization cone and substrate can move straightaway, the intersection point of the spray axis and the substrate can be located at any place of the substrate. The relative motion is repeated and controlled according to the feedback of sensors, through which the materials are accumulated layer by layer. The atomized droplets are spread throughout the substrate surface and the deposits shape at anytime is similar to the end shape. The rate of heat disappearing via the convection of the surface and the conduction of the substrate is faster than that of the traditional spray deposition process, which is attributed to the larger heat conducting area. This advantage becomes more obvious with the increase of the deposits height. It can produce fine microstructure with porosity of about 8% (volume fraction) and yield of about 70%.

The dimensions of as-sprayed 6066 aluminum alloy (Al-1.37Si-1.37Mg-0.77Cu-0.07Mn, mass fraction, %) were d200 mm × 130 mm. The samples were machined into d38 mm × 65 mm for thixoforming. The annealing treatment samples were cut into height of 10 mm × 10 mm × 20 mm from the locations where the microstructures were representative.

2.2 Thixoforming

The prepared samples were heated using 0.5 ℃ grade accuracy electric furnace and hot-pressed using YH41-63C model hydraulic press. The measured solidification temperature zone is 598~ 640 ℃ by DTA. The samples were kept at prefixed temperature for 1 h. The furnace was located near the press to ensure that the samples can be transported into the die in several seconds (about 2 s). The die was preheated to about 260 ℃ to avoid rapid cooling of samples. The inner diameter of upper mould was 40 mm, which could hold the expanded samples. The extrusion ratio was 11∶1. The lower mould of hot pressing had two perpendicular and intersecting rectangle grooves with cross section dimensions of 10 mm (height) and 8 mm (width). The ability of materials to fill the grooves reflected the forming property. All the dies were made from stainless steel. As-pressed articles were water quenched.

2.3 Annealing trials

To evaluate the microstructure change at different temperatures for different annealing times, samples were machined into 10 mm × 10 mm × 20 mm from the representative locations and annealed under different conditions. After water cooling, the cross sections in the half height were chosen to measure with the aim of avoiding influences caused by oxidization and other factors on microstructures.

2.4 Testing

Series of samples in every stage of the whole procedure of the experiment were subjected to comprehensive tests in order to characterize the materials and to assess the effectiveness of the new route. These included inspection by optical microscopy, SEM and radiography; tensile tests for yield strength, UTS and elongation and hardness measurements. Average grain sizes were determined by a linear intercept technique.

3 RESULTS AND DISCUSSION

3.1 Annealing treatment

A series of typical microstructure graphs are shown in Fig. 1. The mean grain size of the deposits is about 16 μm. The fine microstructure result from the great nucleation ratio, shorter staying time at high temperature and impure particles embedded in grain boundaries[10]. There is a small fraction of pores distributed along the equiaxed grains boundaries. After staying at 580 ℃ (18 ℃ below the solid phase line) for 40 min, a slight coarsening from previous 16 μm to current 20 μm occurs. The pores size and total volume fraction increase a bit. The migration of grain boundaries at high temperature by the force of gas/solid interface energy results in combination of pores. The reasons for the increasing porosity may account as: 1) the plastic deformation caused by the elevated gas pressure in closed pores and the dramatic decreasing of material yield strength; 2) absorbed gas during spray deposition escapes to form pores.

This tendency gets more obvious at 610 ℃ (12 ℃ above the solid phase line) for 40 min (as above in Fig. 1(b)). The grains coarsen apparently while pores and porosity increase, which could also be attributed to the same reasons mentioned above. In addition, the presence of liquid phase accelerates solute diffusion, which leads to larger grain coarsening rate (as will be discussed later) and more rapid mergence of pores. With the prolongation of annealing time at 620 ℃, the grains grow bigger and liquid phase gathers in grain boundaries, the pores contract and total porosity decreases. This tendency continues at 630 ℃ and 640 ℃ (shown as Fig. 1(d)). This densification seems like sintering with liquid phase. The driving force was the reduction of interface energy (include gas/solid, gas/liquid and solid/solid interface energies). The grains keep equiaxed during growth, which meets the requirement of thixoforming.

Average grain sizes are illustrated in Fig. 2. The grains grow slowly in the full solid state. There are two possible inhibiting factors: 1) minority high temperature particles (such as nitride and other impurity) at the grain boundaries may prevent boundary migration[9]; 2) fine pores can fix boundaries.

Liquid phase benefits the transportation of solute from large particles to small particles, thus accelerates the procedure of grain growth. The accurate coarsening mechanism of as-sprayed deposits in semisolid state is not established. Further experiments are required to elucidate the mechanisms and kinetics of grain growth in both fully solid state and semisolid state of spray deposits.

3.2 Semisolid extrusion

3.2.1 Forming force

Table 1 shows the force during steady extrusion of as-sprayed 6066 aluminum alloy at different temperature in the same extrusion ratio of 11∶1. The forming force decreases obviously with elevating extrusion temperature. The sample was so irregular that it could not be transferred into extrusion mould successfully at temperature more than 640 ℃. The extrusion at 600 ℃ was not finished because the necessary force exceeded the maximum force of the press (490 MPa). The force used in semisolid extrusion is about 1/3 of that in full solid extrusion (1200 MPa), which shows significant advantage of thixoforming process that there is no need for large-scale pressing equipment, which leads to cost saving.

Table 1 Extrusion force of as-sprayed 6066 Al alloy with same extrusion ratio of 11∶1

Extrusion temperature/℃	Extrusion Force/MPa
480	1200
610	480
620	408
630	380

3.2.2 Microstructure

Significant densification occurred in as-extruded 6066 aluminum alloy with porosity lowered to less

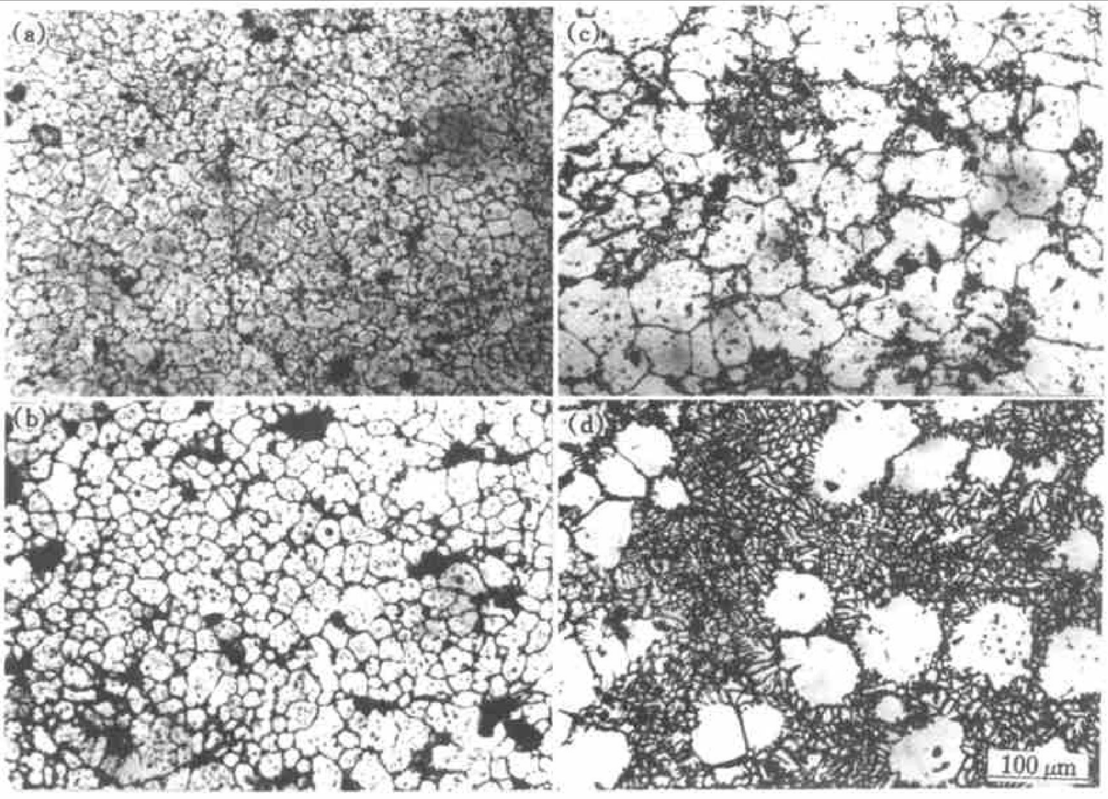

Fig. 1 Annealed microstructures of spray deposited 6066 aluminium alloy
(a) —Spray deposit; (b) —Annealed at 610 ℃ for 40 min; (c) —Annealed at 620 ℃ for 120 min; (d) —Annealed at 640 ℃ for 40 min

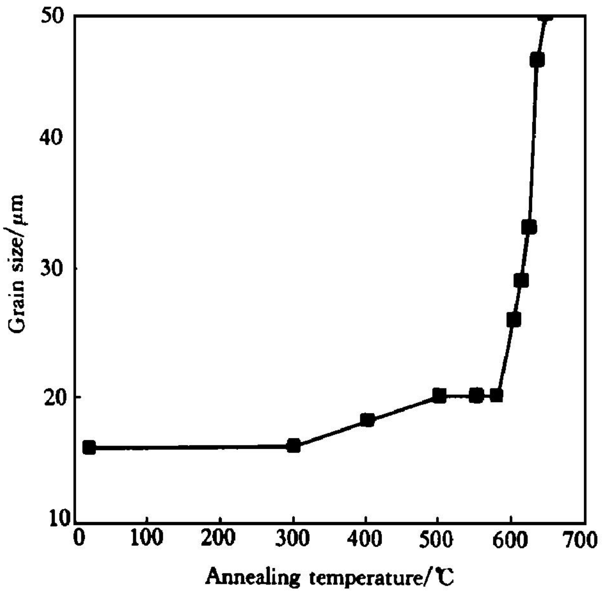

Fig. 2 Average grain sizes of 6066Al annealed at different temperatures for 40 min

than 1%. The residual porosity may result from gas entrapment during extrusion[6] and the former gas pores. The pores are of less than 1 μm in diameter and spheroidal appearance and disperse uniformly in the matrix.

As-extruded samples underwent T6 treatment (solution heat treated at 530 ℃ for 1 h, aging at 175 ℃ for 8 h) and subsequently metallographic inspection, as shown in Fig. 3. The cross section is characterized by fine grains of about 20 μm and the vertical section kept streamline like microstructure. These streamlines are fine and dense, from which the conclusion that there are dense and fine precipitates occurring in extrusion process could be drawn. The precipitates disperse in both grain boundaries and inner grains, as shown in Fig. 4. T6 heat treatment increases as-extruded precipitates size from about 1 μm to about 2 μm and their volume fraction a bit. The fine and dispersed precipitates after extrusion can accelerate solution treatment, and may account partially for no apparent recrystallization occurring in T6 heat treatment. Another reason for inhibiting recrystallization may be the low stored energy generated by deformation at very high hot-working temperature. The distinct microstructure is required to conduct detailed measurements and analysis.

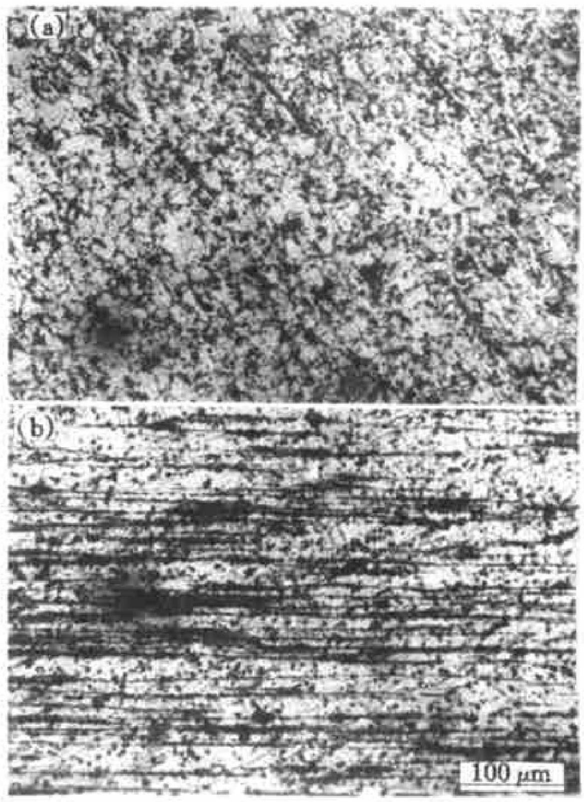

Fig. 3 Metallograph of as-extruded 6066 aluminium alloy at 620 ℃ under T6 condition
(a) —Cross section; (b) —Vertical section

3.2.3 Mechanical properties

The typical properties are lined up in contrast with conventional casting and extrusion process, as shown in Table 2. The strength of as-extruded mate-

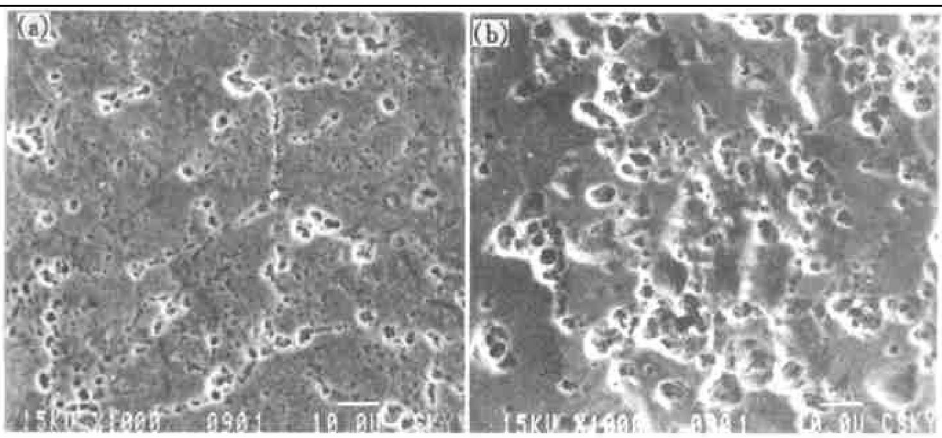

Fig. 4 Appearance of precipitates in as-extruded 6066 aluminium alloy (dark blackzones are the precipitates)
(a) —As-extruded; (b) —T6 condition

rial exceeds that of conventional extruded material, at the same time the elongation improves more outstandingly with an improvement by 100 percent under T6 condition. The typical appearance of fracture evinces excellent plasticity of as-extruded and T6 heat treated 6066 aluminum alloy, as shown in Fig. 5, which may benefit from dispersed fine precipitates.

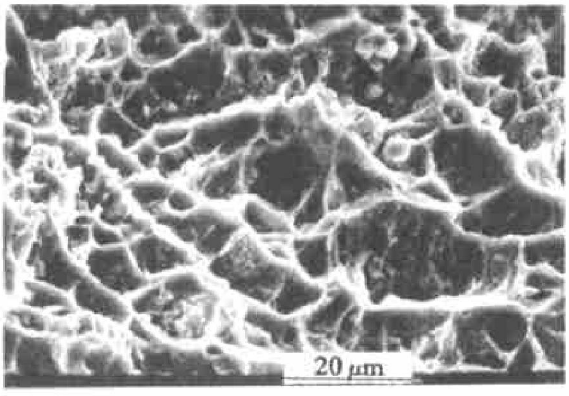

Fig. 5 SEM morphology of fracture of as-extruded and T6 treated 6066Al alloy

3.3 Semi-solid pressing

3.3.1 Formability

The constant forming pressure of 480 MPa was adopted in these experiments. The formability is judged mainly by the appearance of the top cross, and it is dependent on the deformation temperature as expected. In the experiment condition, the acceptable heating temperature of samples should be over 610 ℃ (12 ℃ above the measured solid phase temperature) with incorporated bottom mould temperature of about 260 ℃. By use of proper process parameters, as-produced cross surface is smooth without cracks and obvious pores. Density of cross is the same as that of other sites (2.73 g/cm^3 typically).

3.3.2 Microstructure

Fig. 6 shows typical microstructure of as-pressed 6066 aluminum alloy at 620 ℃ under the condition of no heat treatment, solution treatment and T6 heat treatment respectively. Fig. 6(a) displays little porosity and large equiaxed grains over 100 μm with low melting point phase distributed along the grain boundaries. The grains get larger than those measured in annealed samples. The possible explanation is that most pores are eliminated and many high temperature minority particles are merged by deformed grains under the action of pressure, which weaken the inhibiting factor of the grain boundary migration. The most distinct phenomenon occurred when solution treatment was conducted to as-pressed material, that the grains were fined significantly, seen in Fig. 6(b). Complete recrystallization took place during solution treatment with grain size fined from original about 100 μm to about 25 μm. The driving force is the stored energy generated by plastic deformation and large excess solution due to rapid solidification after pressing. There must be large coring rate during above recrystallization procedure, for which the reasons can be depicted as: 1) quenched liquid phase

Table 2 Typical mechanical properties of 6066 aluminium alloy by two kinds of processes

Process	Heat treatment	Tensile strength/MPa	Yield strength/MPa	Elongation/%
Cast+ extrusion[11]	–	< 200	–	16
Cast+ extrusion[11]	T6	> 345	> 310	8
As-extruded	–	233	150	21
As-extruded	T6	362	313	16

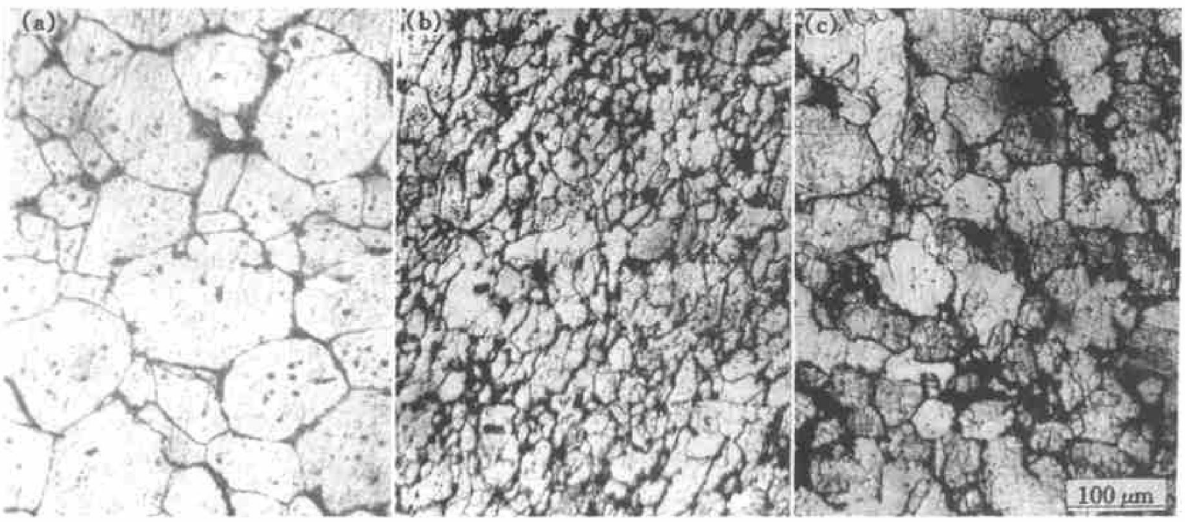

Fig. 6 Typical microstrutures of as-pressed 6066 aluminium alloy at 610 ℃
(a) —As-pressed; (b) —Solution treated; (c) —T6 treated

from semisolid state will decompose when rapidly heated to solution temperature. The reaction production will disperse uniformly with fine size, which supplies fine recrystallization cores; 2) quenched solid particles from low semisolid temperature are of excess solution, thus decompose at subsequent solution temperature which benefits recrystallization; 3) large heating rate increases actual recrystallization temperature, which benefits improvement of recrystallization rate; 4) minority particles such as nitride improve recrystallization coring.

Much more experiments are required to achieve deep comprehension about it.

Fig. 6(c) shows a typical microstructure of as-soluted and subsequently aged at 175 ℃ for 8 h (T6) 6066 aluminum alloy. The grain size seems larger than that in Fig. 6(b), which perhaps rises from some accidental causes. Abundant subgrains in Fig. 6(c) are consistent with that in Fig. 6(b), which may be related to abundant dislocation caused by deformation.

Fig. 7 shows the distribution of low melting point phase in the matrix. EDAX measurement reveals that its composition includes about 21% Si and 20% Mg. Solution section profile of Al-Mg-Si alloys indicates that the solution of Si and Mg at 530 ℃ is less than 0.5% and 1.0% respectively. It indicates that there is large excess solution of Mg and Si in the Al matrix, which benefits recrystallization coring mentioned above. X-ray spectra of different condition samples had agreement with this phenomenon.

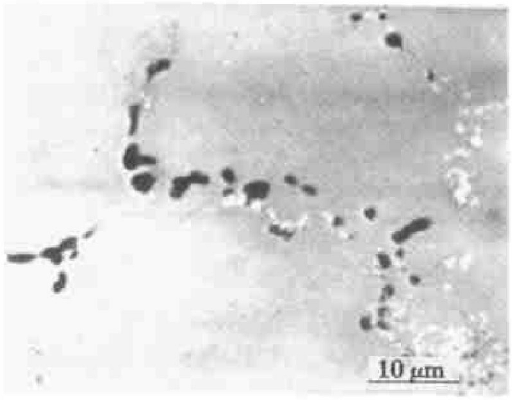

Fig. 7 SEM diagram of as-pressed 6066 aluminum alloy

Fine recrystallization grains in solution treatment state are advantageous to improve the rate of precipitates coring, thus achieves good distribution and size of the precipitates.

3.3.3 Mechanical properties

As-pressed materials were measured in terms of hardness and tensile properties, Fig. 8(a) and Fig. 8(b) give the results respectively. Hardness is obviously lower in solution state than in aging state, which is consistent with conventional situation. Hardness increases sharply with increasing aging time in the initial stage, and attained the peak value for 4~ 5 h. Thereafter it waves slightly near the peak value till aging time of 24h. Hardness seems similar to one another at different thixoforming temperatures. Fig. 8(b) shows the tensile strength results. The tensile strength of 2 h aged sample increases sharply compared with that of as-pressed samples and is similar to those of 8h and 16h aged samples. This result shows good agreement with measured microstructures.

All the measured tensile properties are listed in Table 3 in contrast with conventional forged materials. There exists a slight down slope of elongation when age time exceeds 2 h. Although measured data are not enough to reveal the whole curve of plasticity, we can estimate that the best comprehensive mechanical property is between age time of 2h and 8h.

Compared with semisolid extrusion materials,

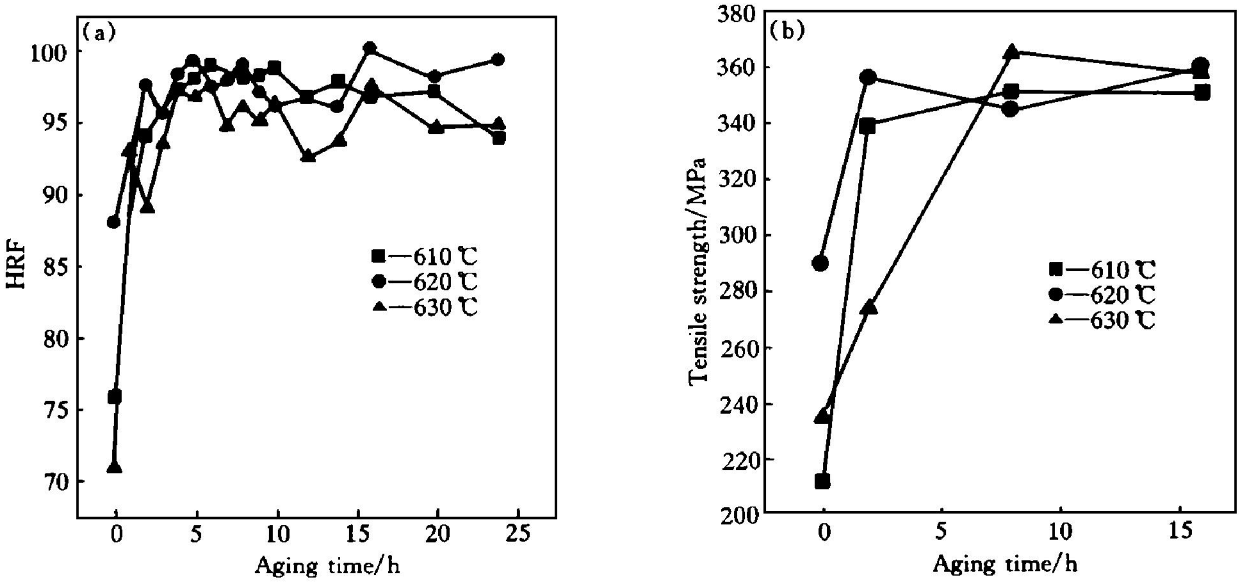

Fig. 8 As-pressed 6066 aluminium alloys in different heat treatment state
(a) —HRF hardness; (b) —Tensile properties

Table 3 As-pressed 6066 aluminium alloy mechanical properties

Material	Forming temperature	Heat treatment	σ_b /MPa	$\sigma_{0.2}$ /MPa	δ /%
Al-1.37Si-1.37Mg-0.77Cu	610	0	212	168	4.5
		Aging for 2h	339	261	4.0
		Aging for 8h	351	300	3.0
		Aging for 16h	351	302	4.2
	620	0	322	260	9.6
		Aging for 2h	356	310	6.7
		Aging for 8h	349	297	4.3
		Aging for 16h	363	308	4.8
	630	0	235	130	13.9
		Aging for 2h	274	208	8.7
		Aging for 8h	365	293	6.5
		Aging for 16h	358	289	6.0
Al-1.4Si-1.1Mg-1.0Cu-0.8Mn[11]	Cast+forge	0	150	83	18
		T6	395	359	12

as-pressed materials had relatively low plasticity, which may be attributed to large extent to different distributions and sizes of precipitates.

4 REMARKS

This study indicates that a new processing route combining spray deposition with thixoforming is viable in terms of technology. On the other hand, this route possesses cost saving potential as

1) High production rate of spray deposition and avoidance of additional long time stirring necessary in semisolid state.

2) Short peak aging time due to very fine grains in solution state.

3) High material utilization ratio for production of complex components due to one-step shaping without subsequent machining.

In addition, this route can realize automation just like other thixoforming process.

5 CONCLUSIONS

1) 6066 aluminum alloy microstructures produced by multi-layer spray deposition show insignificant grain size coarsening in the solid state below the solid phase line temperature.

2) Grains of multi-layer spray deposited 6066 aluminum alloy in semisolid state maintain equiaxed, with low melting point phase liquid separating solid grains.

3) Semi-solid multi-layer spray deposited 6066 aluminum alloy can be extruded under pressure of 480 MPa, about 1/3 of that under conventional hot extrusion. As-extruded material shows excellent distribution and size (about < 1 μm) of precipitates. There exists a slight coarsening from less than 1 μm to about 2 μm under T6 condition for these particles. No obvious recrystallization occurs during T6 treatment.

4) Compared with conventional casting, as-extruded 6066 aluminum alloy has similar tensile strength while 30% ~ 100% improvement in plasticity, and complex 6066 alloy articles can be formed by pressing under about 480 MPa.

5) Fine recrystallization grains (about 25 μm) are achieved after solution treatment in as-pressed 6066 aluminum alloy, resulting in decrease in peak aging time by half (about 4h).

6) As-pressed 6066 aluminum alloy mechanical properties are similar to that by conventional forging, with slight decrease in plasticity.

[REFERENCES]

[1] Flemings M C, Rlek R G and Young K P. Rheocasting [J]. Mater Sci Eng, 1976, 25: 103-117.

[2] Kirkwood D H. Semisolid metal processing [J]. Inter Mater Rev, 1994, 39(5): 173-189.

[3] Flemings M C. Behavior of metal alloys in the semisolid state [J]. Metall Trans, 1991, 22A: 957-978.

[4] Leatham A G. Spray forming: alloys, products and markets [J]. MPR, 1999, 5: 28-37.

[5] Ward P J, Atkinson H V, Anderson P R G, et al. Semi-solid processing of novel MMCs based on hypereutectic aluminium-silicon alloys [J]. Acta Mater, 1996, 44(5): 1717-1728.

[6] McLelland A R A, Atkinson H V, Kapranos P, et al. Thixoforming spray-formed aluminium/silicon carbide metal matrix composites [J]. Mater Lett, 1991, 11(1): 26-30.

[7] Valer Goni J, Rodriguez-Ibabe J M and Urcola J J. Strength and toughness of semisolid processed hypereutectic Al/Si alloys [J]. Scripta Materialia, 1996, 34(3): 483-489.

[8] CHEN Zhen-hua, JIANG Xiang-yang and YANG Fu-liang, et al. Multi-layer spray deposition technology [J]. The Chinese Journal of Nonferrous Metals, (in Chinese), 1995, 5(4): 66-72.

[9] ZHANG Hao. Study on multi-layer spray deposition principle and materials [D]. Central South University of Technology, (in Chinese), 1998.

[10] CHENG C, Annavarapu S and Doherty R. Modeling-based microstructure control in spray casting [A]. Proc of 2nd Int Conf on Spray-Forming [C]. Swansea, UK: Woodhead Pub Ltd, 1993. 67-83.

[11] ASM. Metals Handbook [M], Vol. 2, 9th edition. Beijing: Mechanical Industry Press, 1994. 168.

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