Coaxial jet melt-spinning of glassy alloy ribbons

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A chill block melt-spinning process is described for the manufacture of continuous metallic ribbons of uniform geometry. Ribbons wider than those obtained by conventional melt-spinning in air are formed by stabilization of the molten alloy puddle through the use of a gas stream coaxial with the molten alloy jet impinging on the moving substrate surface. The dependence of ribbon geometry on process parameters for coaxial jet casting is similar to that found in conventional chill block melt-spinning except that somewhat greater molten alloy spreading may occur in the former process. Stress relaxation and the surface geometry of ribbons cast using the coaxial jet technique is found to be similar to that of vacuum and helium-cast samples having the same dimensions.

1. Introduction

Several variations of chill block melt-spinning (CBMS) are described in the literature with regard to the manufacture of metallic ribbons directly from the melt [1–6]. In the basic process, a molten alloy jet is first formed and then impinged onto a rapidly moving substrate surface, resulting in the formation of a molten alloy puddle from which ribbon is continuously formed and chilled. Perturbation of the melt puddle may arise from its interaction with the gas boundary layer established on the moving substrate surface. The results of such melt puddle perturbation include the possible occurrence of surface texture and voids as well as edge serrations in the melt-spun ribbon. Such geometric asperities have been shown to result in a degradation of the magnetic properties [7]. As has been discussed elsewhere [8], processing limitations set by the gas boundary layer established on the rapidly moving substrate surface may be overcome by casting in partial vacuum or in a controlled atmosphere. Attempts to eliminate the gas boundary layer by the use of a mechanical wiping system or similar devices acting on the substrate surface behind the point of melt jet impingement have been ineffective because of the short distance over which the gas boundary is re-established. [9].

Besides elimination or modification of the gas boundary layer, improved ribbon geometry should also be obtainable through stabilization of the melt puddle against perturbation. Casting techniques described by Narasimhan [10] and by Takayama and Oi [11] essentially rely on a small crucible–substrate spacing to offer a mechanical constraint of the molten alloy against perturbation. Close geometric tolerances are required by such techniques throughout the course of a casting run in order to obtain a sample of uniform geometry. The method of ribbon manufacture described in the current work is a technique in which both melt puddle and boundary layer control are simultaneously exercised.

The coaxial jet melt-spinning (CJMS) technique is based on a stream of gas acting on the melt puddle, stabilizing it against perturbation while simultaneously disrupting the gas boundary layer on the moving substrate surface during casting.

2. Coaxial jet casting

As is implied by the name, coaxial jet casting refers to a chill block melt-spinning process in which a gas stream flows coaxially with the melt jet to bear down on and stabilize the molten alloy puddle against the perturbing effects of the gas boundary layer on the rapidly moving substrate surface. An example of a coaxial casting apparatus is schematically shown in Fig. 1. The bearing gas

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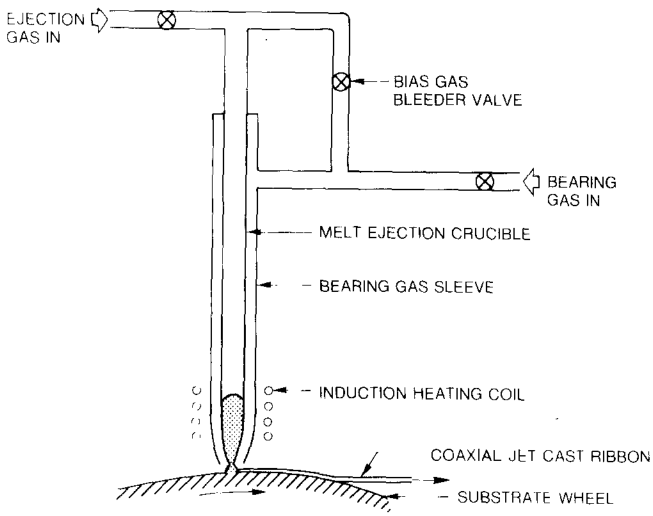

Figure 1 The CJMS system. A gas stream bears down on the melt puddle and stabilizes it against perturbations which could result in geometric asperities in the ribbon product.

sleeve and nozzle are designed so that gas flow is directed at the periphery of the melt puddle from which ribbon is formed, the minimum amount of gas flow required for smooth ribbon geometry being empirically-established. Because of the small coaxial crucible–substrate surface spacing (typically <5 mm), the bearing gas flow may create a high-pressure region just outside the orifice of the melt ejection crucible. The resulting tendency for the molten alloy column to rise out of the heating zone prior to casting is counteracted by the bias gas bleeder circuit, shown in Fig. 1. The dynamics of ribbon formation in coaxial jet casting seem similar to that in conventional CBMS.

The nature of CJMS may be understood by the investigation of ribbons formed under various process conditions, as summarized in Table I. A schematic diagram showing the variables listed is shown in Fig. 2. Ribbon width and thickness, as measured by micrometer, are given by w^{mic} and t^{mic}, respectively. Average ribbon thickness calculated from mass divided by length, density and width is given by $\bar{t}$. Sample C3 is shown in Fig. 3a and was made using conventional CBMS in air. Argon was used as the melt ejection gas and OFHC copper was used as the substrate material. As predicted by relations appearing elsewhere [8], the gas boundary layer Reynolds number of approximately 8400 for the sample shown is greater than the empirically-defined critical value of approximately 2000 and therefore explains the occurrence of fine edge serrations along the length of the ribbon. This type of geometric asperity is

TABLE I Summary of CJMS and CBMS experiments. The variables listed in column headings are illustrated in Fig. 2.

Sample number	ϕ (mm)	y (mm)	δ (mm)	α (°)	P_{ej} (kPa)	P_b (kPa)	v_r (m sec^{-1})	w^{mic} (mm)	t^{mic} (μm)	$\bar{t}$ (μm)
C1	0.76	4	2	90	340	14	36.65	3.8	38	32.1
C2	0.76	4	2	90	210	14	36.65	–	~35	–
C3	0.76	4	–	90	210	–	36.65	–	~30	–
C4	0.76	4	–	90	270	–	36.65	3.7	33	29.6
C5	0.76	4	–	90	270	–	36.65	3.9	38	28.1
C6	0.51	9	5	85	310	27	52.36	2.8	25	19.8
C7	0.51	9	5	85	310	27	39.27	3.1	28	24.8
C8	0.51	9	5	85	310	27	26.18	3.5	45	34.0
C9	0.51	9	5	85	310	27	18.32	3,8	~60	43.5
C10	0.51	9	5	70	310	27	39.27	3.0	35	25.8
C11	0.51	4	4	90	310	14	39.90	2.0	24	–
C12	0.51	4	2	90	310	14	39.90	2.0	27	–
C13	0.51	4	1	90	310	14	39.90	2.1	25	–

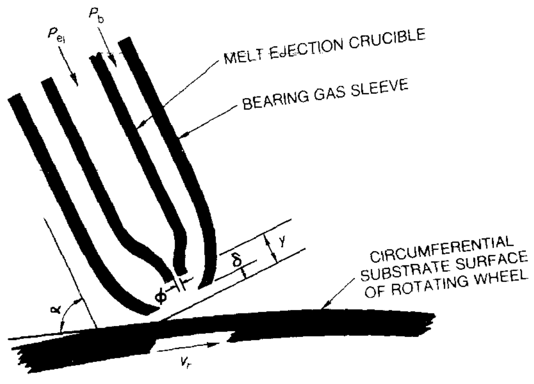

Figure 2 Identification of the variables appearing in Table I.

typical when casting ribbons wider than between 1 and 2 mm in air. A schematic representation illustrating CBMS at such a hypercritical gas boundary layer Reynolds number is shown in Fig. 4a.

Sample C1 in Fig. 3b was made using the CJMS method using air as the bearing gas. The substantial improvement in ribbon geometry may be attributed to enhanced melt puddle stability. Fig. 4b is a schematic diagram showing the nature of ribbon formation when conducting coaxial casting under such processing conditions. Sample C2 in Fig. 3c is shown as an example of a ribbon made under improper coaxial jet casting conditions. Processing was identical to that in the previous run except that the melt ejection argon pressure was set at 210 kPa rather than at 340 kPa. The ribbon sample shows evidence that severe melt puddle disruption must have occurred, resulting in the poor geometry of the specimen shown. It is presumed that the lower melt ejection pressure used resulted in a narrower melt puddle, and that the bearing gas flow impacted on the moving substrate surface just outside the puddle, as shown schematically in Fig. 4c, causing extensive melt puddle perturbation. The vacuum-cast (C4) and helium-cast (C5) geometric equivalents of coaxial jet cast sample C1 are shown in Fig. 3d and e, respectively. Note that the fluid dynamics of the molten alloy during casting in each of these three samples must have been similar, based on a comparison of ribbon dimensions given in Table I and on the occurrence of slight ribbon edge shoulders in each of the samples.

As has been indicated in the preceding description of the experiments, the technique of CJMS must be used properly in order to obtain rapidly-quenched ribbons of uniform geometry. The most important condition seems to be that the bearing gas flow be made to impact atop the periphery of the melt puddle in order to promote stabilization against gas boundary layer perturbations. This means that for a given composite crucible geometry, processing conditions must be adjusted to produce ribbons of width sufficiently great that the condition in Fig. 4b prevails rather than the condition in Fig. 4c. Besides this lower limit to

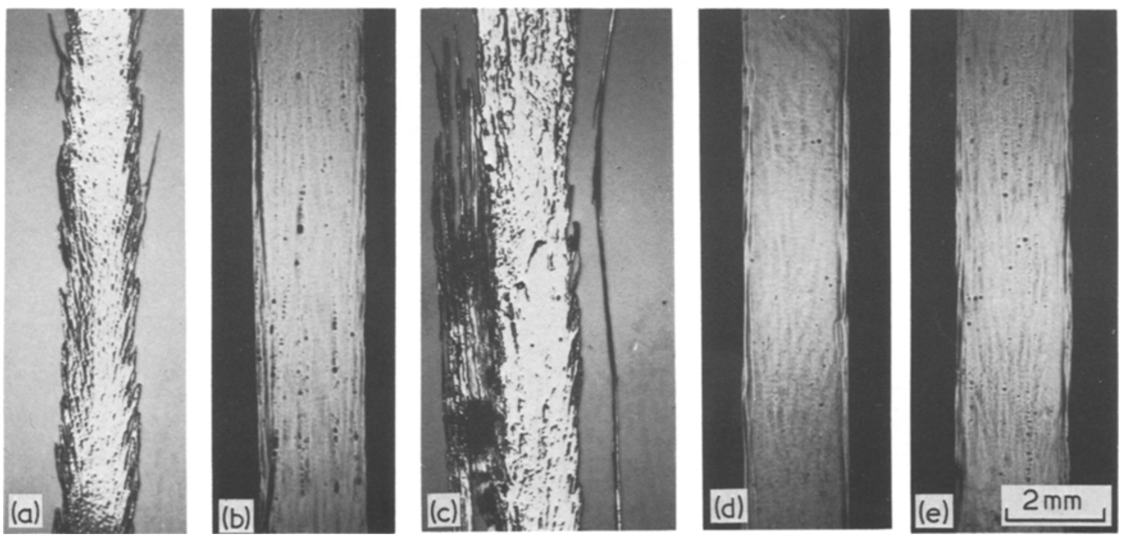

Figure 3 Micrographs of upper surfaces: (a) conventional CBMS in air (sample C3); (b) coaxial jet casting under proper conditions (sample C1); (c) coaxial jet casting under improper conditions (sample C2); (d) vacuum-cast CBMS geometric equivalent of C1 (sample C4); (e) helium-cast CBMS equivalent of sample C1 (sample C5).

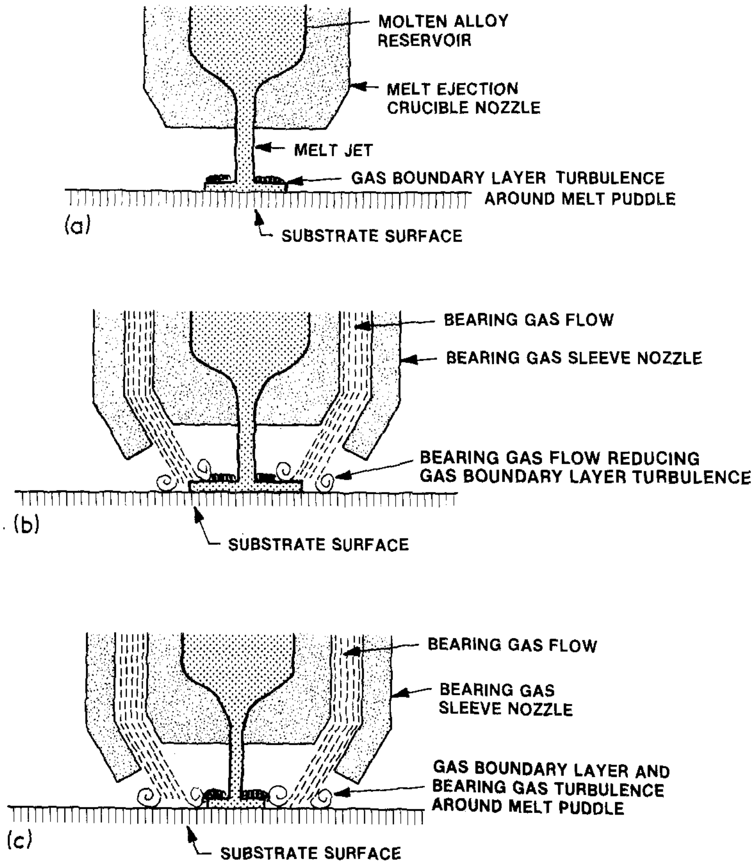

Figure 4 Schematic diagrams showing chill block melt-spinning with the casting direction out of the page: (a) conventional CBMS in air (sample C3); (b) CJMS under proper conditions (sample C1); (c) CJMS under improper conditions (sample C2).

ribbon width set by gas flow effects, there is also a fluid dynamic upper limit to ribbon width which can be made with a single round orifice in the melt ejection crucible using either the coaxial jet technique or conventional CBMS. The maximum width of geometrically uniform ribbon manufactured by using a single round orifice appears to be controlled by the hydraulic jump phenomenon discussed in [12]. Exceeding this empirically-defined maximum ribbon width will result in a ribbon with edge "shoulders" [6], as is seen on some of the samples shown in Fig. 3. The geometric details of the composite nozzle are relatively unimportant in determining whether or not a given coaxial jet design will prove effective. For example, no differences in sample geometry have been measured for ribbons made with a composite nozzle-substrate spacing of $1\,mm \leqslant \delta \leqslant 4\,mm$ (samples C11–C13), which is a clear advantage over a continuous casting process controlled by the crucible–substrate spacing [10, 11] because of the relative insensitivity of process effectiveness on this spacing. As shall be described in the following section, the dependence of coaxial jet melt-spun ribbon geometry on processing parameters is similar to that found for conventional CBMS [6].

3. Effects of process parameters on the geometry of coaxial jet cast ribbon

Experimental results showing the dependence of coaxial jet cast ribbon geometry on variations in several process parameters are shown in Table I. A simple substitution of the data into equations predicting ribbon width and thickness [6] reveals that CJMS ribbon geometry may be approximately described by conventional CBMS geometry relations. There is, however, a rather unique feature of coaxial jet casting. As the substate velocity is decreased, there is an increasing discrepancy between the observed ribbon dimensions and those predicted in [6], as shown in Fig. 5. It is seen that CJMS ribbon ratio w/t is consistently greater than the equivalent CBMS ratio w/t calculated, especially at lower substrate velocities. This is presumably because the melt puddle is taller at low substrate

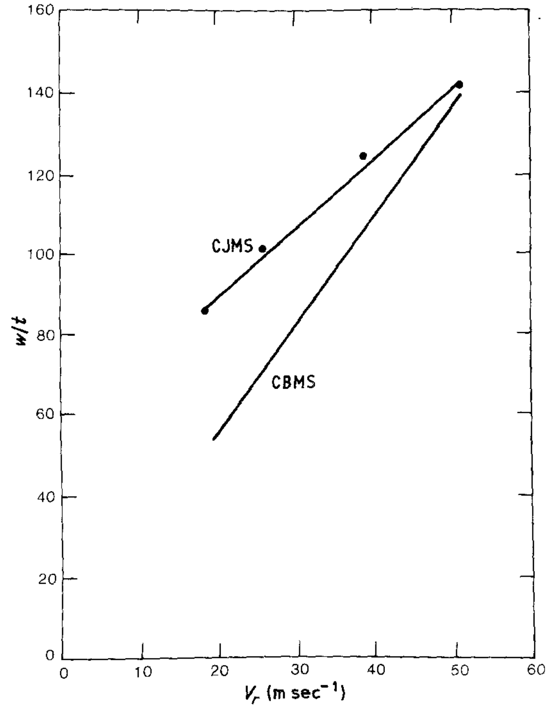

Figure 5 CJMS and typical CBMS ratio w/t against v_r plot for a flow rate $Q \simeq 3\,\mathrm{cm^3\,sec^{-1}}$. Note that the apparent melt puddle spreading caused by the bearing gas flow is enhanced at lower substrate velocities.

velocities. during conventional CBMS. Thus, it is conceivable that the bearing gas pressure is sufficiently great to "flatten out" the melt puddle to a greater extent than would occur under normal CBMS conditions, resulting in a wider, thinner product and accounting for the apparent discrepancy observed. Thus, additional control of the coaxial jet cast sample geometry may be gained by optimizing the bearing gas pressure. Working with the same material flow rate, $v_r = 36.65\,\mathrm{m\,sec^{-1}}$, there is apparently no significant difference in geometry between samples cast by the coaxial method in air and samples cast in either vacuum or helium, as shown by samples C1, C4, and C5 listed in Table I and shown in Fig. 3. The ribbon edge shoulders shown are a consequence of the particular casting conditions used and occur for the same reasons in coaxial jet cast samples as in ribbons made by conventional CBMS. Details of the composite crucible nozzle geometry are shown to be non-critical for samples C11–C13, in which the bearing gas sleeve–substrate surface is changed for a fixed melt ejection crucible–substrate surface distance. Successful operation of the coaxial jet casting system lies in empirically establishing spacings y and δ for a given individual composite crucible nozzle geometry.

TABLE II Results of stress relaxation experiments for glassy $Fe_{40}Ni_{40}B_{20}$ ribbons using various casting methods and substrate wheel diameters. Fixed sample dimensions and a linear substrate velocity were used.

Sample number	Casting method	Wheel diameter (cm)	Stress relaxation
C1	CJMS in air	25.4	0.36
C3	CBMS in air	25.4	0.30
C4	CBMS in vacuum	12.5	0.62
C5	CBMS in helium	12.5	0.56
A1	CBMS in air	12.5	0.63
A2	CBMS in air	7.5	0.64
A3	CBMS in air	2.5	0.76

4. Stress relaxation experiments

Samples C1, C3, C4 and C5 and other glassy alloy ribbons not listed in Table I were subjected to stress relaxation measurements after annealing for 2 h at 500 K in 7.3 mm diameter aluminium cans. Results of these experiments are shown in Table II and in Fig. 6. All ribbons were made using a fixed

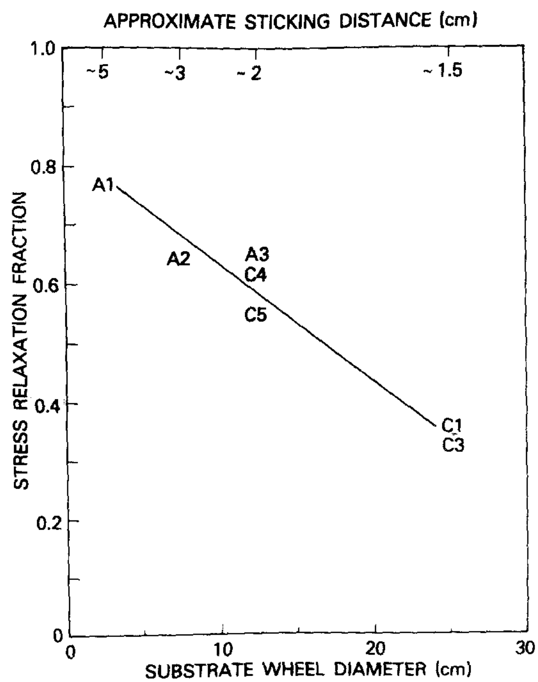

Figure 6 Stress relaxation as a function of substrate wheel diameter for glassy $Fe_{40}Ni_{40}B_{20}$ ribbon samples listed in Table II.

linear substrate velocity $v_r = 36.65$ m sec^{-1} and had thickness about 33 μm and width about 3.8 mm (measured using a micrometer). The scatter of data for the two larger substrate wheels indicates no significant difference in stress relaxation between ribbons of similar dimensions made by CJMS and by CBMS when using a fixed linear substrate velocity.

The increased stress relaxation found when casting glassy alloy ribbon using smaller substrate wheel diameters indicating improved quench rate is presumably related to the increased ribbon–substrate sticking distance, d_s, observed during casting. For the present experiments using approximately 5 g molten alloy per run, the approximate "steady state" ribbon substrate contact distance was found to vary approximately with the inverse square root of substrate wheel diameter (Fig. 6). This observed increase in sticking distance with decreasing wheel diameter is not consistent with earlier observations [14] and is contrary to expectations based on increased centrifugal force with decreasing wheel diameter. Thus, the forces holding the ribbon to the wheel must increase rapidly with decreasing wheel wear or roughening and/or higher substrate temperatures at small diameters.

5. Conclusions

The coaxial jet method for chill block melt-spinning metallic ribbons has been shown to offer greater flexibility than other CBMS techniques based on a free molten alloy jet impinging on a moving substrate. Ribbons of geometry and quench rate equivalent to those made in vacuum or helium are produced at lower cost by using air as the bearing gas acting on the molten alloy puddle. Although the experiments described in the current work pertain to the use of a round melt jet, the use of a planar jet coaxial casting system has been marginally successful in preliminary attempts to manufacture glassy alloy ribbons in air of width greater than 1 cm. Stress relaxation results indicate no significant differences between geometrically equivalent CJMS and CBMS glassy alloy ribbon samples made in vacuum and in helium. This indicates that the ribbon–substrate contact time is a more important factor in determining average quench rate than the atmosphere in which casting is conducted.

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