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Wetting pattern characterization of rapidly solidified centrifuge melt-spun metallic ribbons

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The wetting pattern characterization of rapidly solidified centrifuge melt-spun metallic Al–Ge ribbons has been enabled by measuring the roughness of the rim-contact side of the ribbons. Roughness parameter values were obtained using the SEM ability of amplitude modulation and line scanning of the smooth ribbon surfaces, and micro-computerized digitizing of the line profiles. In such a manner, quantification of the wetting patterns is possible. As a result, the respective contribution of the ejection pressure and the extraction velocity on the wetting patterns of the solidifying ribbons is evaluated. In centrifuge melt spinning, the roughness of the rim-contact surface of the ribbons increases when extraction velocities are increased. Despite the additional roughness and the "bad" thermal contact conditions for effective heat transfer, higher cooling rates (as measured by secondary dendritic arm spacings), are achieved. It appears that a kind of dragging occurs, reducing the ribbon thickness by a shear drawing mechanism. This behaviour is characteristic of centrifuge melt spinning because of the specific hydraulic and heat transfer features involved with the counter-rotating of the crucible and the substrate.

1. Introduction

A new technique for the production of rapidly solidified metallic ribbons, centrifuge melt spinning (CMS), has been presented recently [1–3]. The specific features of CMS are as follows: a rotating crucible which contains the molten alloy is used; due to the centrifugal force, the alloy is expelled from the crucible to solidify on the surface of the rotating substrate; the cooling substrate is a copper rim which rotates in the opposite direction; as the alloy stream impacts on the rim, a molten puddle is formed, from which the rapidly solidified ribbon is extracted, as a result of the counter-rotation of the crucible and the substrate. For both conventional melt-spinning and CMS, the wetting of the liquid metal on the casting rim surface is an important process factor, insofar as it influences the heat transfer at the metal–rim interface and the smoothness of the rim-side surface of the ribbon. The degree of uniformity in mechanical and physical properties along the length of the rapidly solidified ribbons is closely related to the wetting characteristics. Previous work [4] has shown that rim surface conditions do indeed affect the wetting pattern. As an air boundary layer forms in the gap between the crucible and the rim surface, air may be entrapped in the solidifying melt, and air pockets appear on the rim side ribbon surface. This leads to a specific wetting pattern, representing the metal–rim interfacial characteristics. It has been shown [4], that casting performed on a matte substrate rather than on a smooth one, does improve the average heat-transfer coefficient at the metal–rim interface. In CMS, the ejection pressure of the melt can be varied, and the extraction velocity can attain large values (up to $100\,\mathrm{m\,sec^{-1}}$). Cooling rates as high as $10^8\,\mathrm{K\,sec^{-1}}$ are achieved [1–3], due to improved thermal contact of the molten alloy puddle to the cooling substrate, and to a beneficial combination of crucible and substrate velocities.

A matter yet to be investigated, however, is the respective contribution of the ejection pressure and the extraction velocity on the wetting pattern of the solidifying ribbons, and therefore on the heat transfer characteristics at the metal–rim interface. For that purpose, a quantitative study of the wetting pattern is needed, in addition to a comprehensive understanding of the fluid dynamics and heat transfer behaviour of the melt–substrate system [5].

This paper is mainly aimed to present an innovative procedure for performing quantitative analysis of the wetting patterns of melt-spun metallic ribbons. The roughness of the rim-contact surface has to be measured, as a representative parameter resulting from the specific casting conditions. Roughness parameters are in good correlation with actual quenching rates.

2. Experimental procedures and results

The binary hypoeutectoid alloy in an as-received Al–12 at. % Ge composition was processed by CMS, as described previously [1–3]. All rapidly solidified Al–Ge ribbons were microcrystalline, exhibiting a dendritic or degenerate-dendritic microstructure. Extended metastable solid solubility of germanium in aluminium (up to 7.7 at. %) has been obtained [6]. Ejection pressures ranged from 1.8 to 269 kPa (corresponding to crucible rotating at linear velocities from 6.5 up to $25\,\mathrm{m\,sec^{-1}}$), and substrate velocities ranged from nil to $78\,\mathrm{m\,sec^{-1}}$. Volumetric flow rates were

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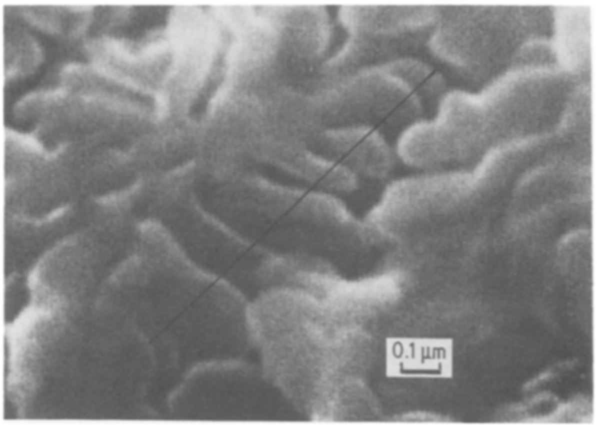

Figure 1 DAS, 15 μm thickness ribbon; ejection pressure = 97 kPa, rim velocity = 78.9 m sec^{-1}.

from 1.3 up to 5.0 cm^3 sec^{-1}. Ribbons were produced, at various thicknesses (15 to 87 μm) and widths (0.93 to 3.23 mm). Secondary dendritic arm spacings (DAS) were measured using scanning electron microscope (SEM) examination. It is generally admitted that dendrite size measurements can provide information on the cooling history [7]. The highest experienced cooling rate, measured on a 15 μm ribbon was $\approx 10^8$ K sec^{-1}, corresponding to a secondary dendritic arm spacing of ≈ 0.11 μm (Fig. 1). Electron microscope examination of the wetting pattern of the rim-contact side of the ribbons shows that, for the same total (crucible + rim) linear velocity ($V_{\mathrm{tot}} = 40$ m sec^{-1}), and different ejection pressures $P_{\mathrm{ej1}} = 1.8$ kPa and $P_{\mathrm{ej2}} = 269$ kPa, the wetting pattern is finer when using higher pressures (Fig. 2). When comparing wetting patterns on the rim-contact side of two ribbons, cast at the same ejection pressure ($P_{\mathrm{ej}} = 97$ kPa) and different substrate velocities ($V_{\mathrm{sub1}} = 5.8$ m sec^{-1} and $V_{\mathrm{sub2}} = 39.4$ m sec^{-1}), it appears that increasing the substrate velocity yields a smoother wetting pattern (Fig. 3). Quantification of the ribbon rim-contact surface appearance is necessary, in order to enable proper correlation between the roughness of the surfaces and the various process parameters.

Ribbons were therefore examined by SEM, using the amplitude-modulated line scanning ability of the microscope which allows observation of Y modulated scanned images. The vertical deflection signal of the CRT is modulated by the video signal, and a Y-modulation image is thereby obtained. The modulation amplitude can be adjusted. Amplitude modulation is used together with line scanning. A horizontal line scan is performed on the ribbon's width and yields a line profile. Such a line profile represents the topographical features of the scanned surface. Line scan is repeated at a vertical repetition speed of 7 scan lines per second. When all scanning and photographic parameters (objective lens aperture, amplitude, contrast and brightness of secondary electron image and camera) are appropriately set, polaroid micrographs are obtained, as shown in Fig. 4 (taken with a 25 keV Jeol scanning electron microscope, operating at 120 μA current and using PN 52 polaroid film for the micrographs). Fig. 4a is a × 60 magnification of the ribbon shown in Fig. 1, where the line profiles are clearly seen on the ribbon surface (rim-contact). Fig. 4b is another scan of the same surface, taken at higher magnification, × 240. Fig. 4c, still from the same ribbon, is the micrograph used for further analysis (digitizing). It represents a scan performed on the same ribbon at the × 240 magnification, without the ribbon at the image's background.

The image analysis of the line profiles (as seen in Fig. 4c) is performed by: (i) photocopying the micrographs to produce transparancies, where the profile lines are black on a transparant background; (ii) scanning the transparancies, using a high-resolution digitizer (Thunderscan$^{\mathrm{TM}}$), connected to a microcomputer (Macintosh$^{\mathrm{TM}}$); (iii) projecting the processed profile lines on the computer's screen. Fig. 5 shows typical line profiles, as they appear on the screen. Magnification of selected areas in Fig. 5 can be obtained using the "Fatbits" ability of the microcomputer software (MacPaint$^{\mathrm{TM}}$); (iv) evaluating the roughness of each profile line, using an original software aimed at that purpose (written in Forth language), which computes the root-mean-square of all values of the roughness profile within the measuring length. The resolution of the r.m.s. computation is one pixel; (v) repeating the roughness evaluation of ten different profile lines, taken at different sections of one specific ribbon, in order to obtain the roughness parameter of the ribbon, together with its standard deviation.

Table I shows typical roughness (values rounded at 0.05, with standard deviations) and process

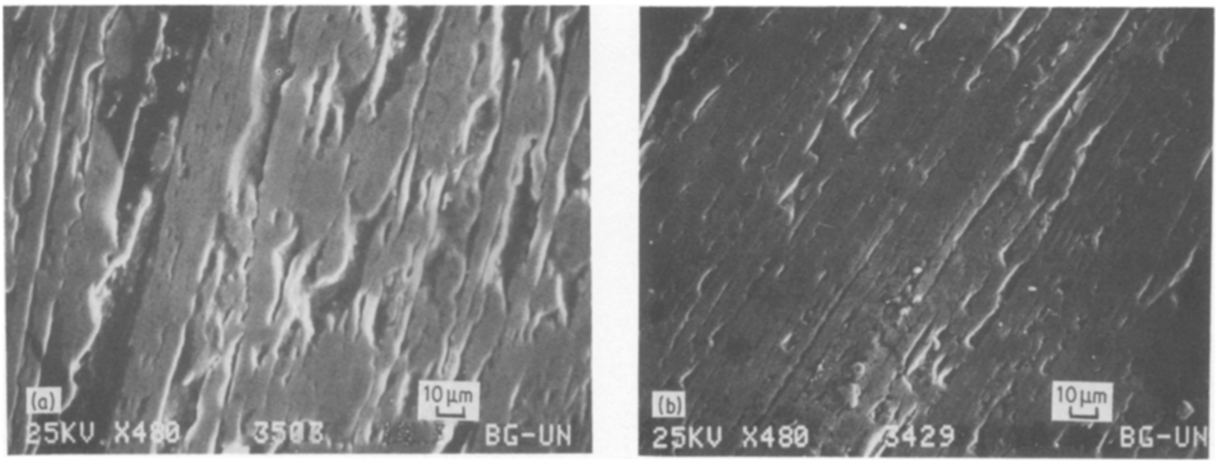

Figure 2 Influence of P_{ej} on the ribbon wetting pattern. $V_{\mathrm{tot}} = 40$ msec^{-1}. (a) $P_{\mathrm{ej1}} = 1.8$ kPa, (b) $P_{\mathrm{ej2}} = 269$ kPa.

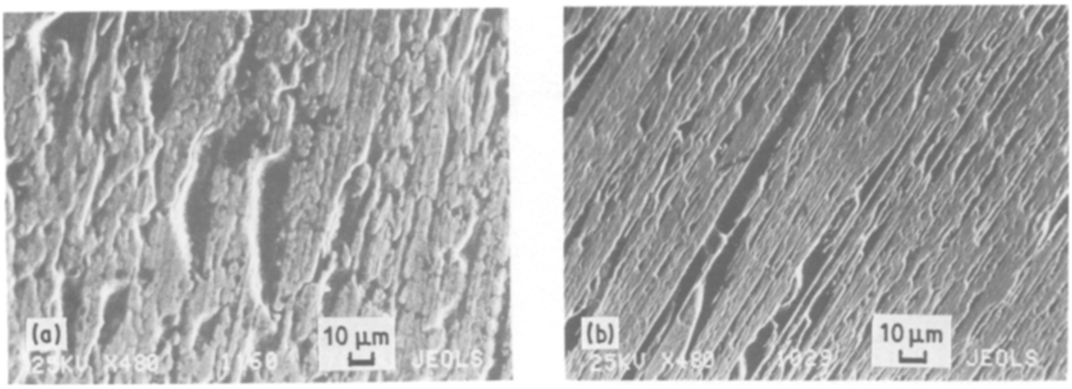

Figure 3 Influence of V_{sub} on the ribbon wetting pattern. $P_{\mathrm{ej}} = 97\,\mathrm{kPa}$. (a) $V_{\mathrm{sub1}} = 5.8\,\mathrm{m\,sec^{-1}}$, (b) $V_{\mathrm{sub2}} = 39.4\,\mathrm{m\,sec^{-1}}$.

parameters as measured on various centrifuge melt-spun Al–Ge ribbons. The ejection pressures and total velocities are as in [3]. Thicknesses and width are average, ±15% values (100 thickness and 20 width measurements for each ribbon). The cooling rates developed on the rim-contact side of the ribbons have been calculated from the dendrite arm spacing measurements using the experimental relationship, for Al–Cu and Al–Si alloys [8]:

$$\lambda \varepsilon^{1/3} = 50\,\mu\mathrm{m}\,(\mathrm{K\,sec^{-1}})^{1/3}$$

where λ is the measured DAS and ε is the cooling rate. DAS values are as evaluated on SEM micrographs.

Average cooling rates $\mathrm{d}T/\mathrm{d}t$ have been evaluated according to:

$$\mathrm{d}T/\mathrm{d}t = [h\Delta T]/[c_{\mathrm{L}}\,th]$$

where h is the heat transfer coefficient (one h value for each ejection pressure), extrapolated from the ribbon thickness dependence on the contact time of the solidifying melt on the substrate and assuming pure heat transfer effects only, with no mechanical dragging of the ribbon out of the melt puddle. The various h values, as calculated by Baram [9], range from 1.56×10^6 to $4.33 \times 10^6\,\mathrm{W\,m^{-2}\,K^{-1}}$, ΔT is the temperature drop (800 K, consecutive to a 150 K melt superheat), c_{L} is the specific heat of the Al–Ge alloy ($3.456 \times 10^6\,\mathrm{J\,m^{-3}\,K^{-1}}$), th is the ribbon thickness.

As shown from the results in Table I, (i) at a given ejection pressure, roughness parameter values do increase with increasing total (extraction) velocity, whatever is the ejection pressure; DAS values decrease systematically, while the computed average cooling rate increases; (ii) at the same total (extraction) velocities, the roughness parameter values are generally lower for higher ejection pressures; (iii) beyond these two previous trends, there is no correlation whatsoever in CMS between the roughness parameter values and either DAS values or average cooling rates.

3. Discussion

The wetting pattern analysis presented here is still in its experimental stage. The exact relationship between peaks and valleys of the digitized image and the real scale topographical features of the ribbon surface

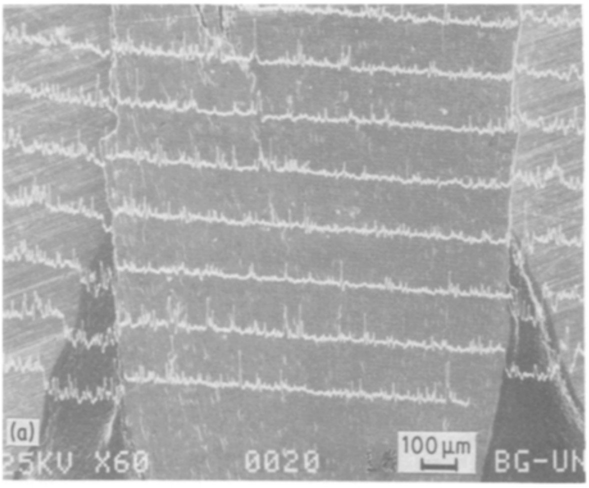

Figure 4 Line scan images. (a) ×60, scan superimposed on the ribbon's image. (b) ×240, scan superimposed on the ribbon's image. (c) ×240, image for further analysis (digitizing).

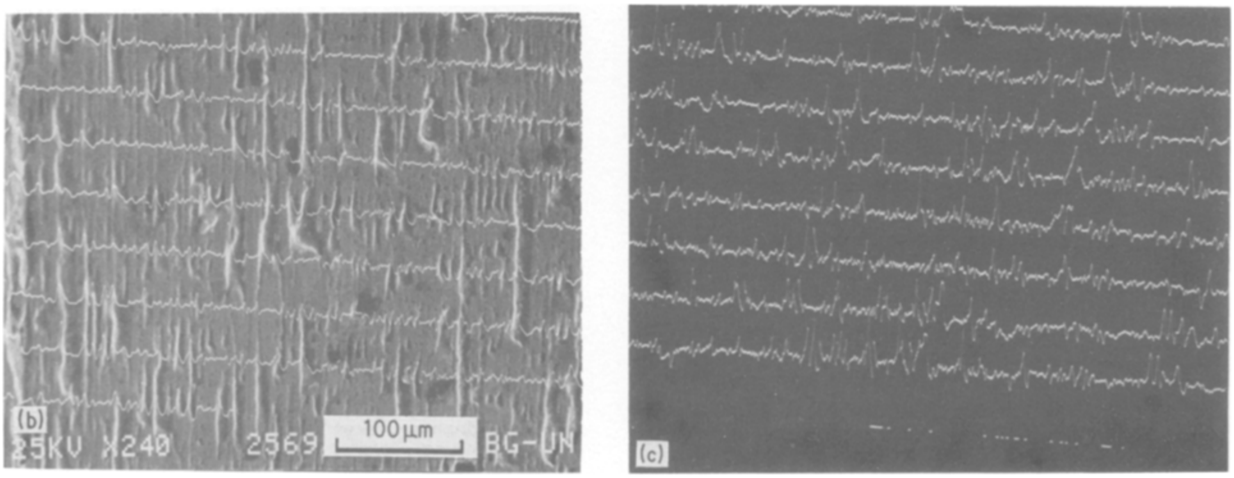

TABLE I Typical roughness and process parameters

Spec. no.	P_{ej} (kPa)	V_{total} (m sec^{-1})	R parameter	DAS (μm)	Thickness (μm)	Width (mm)	Aver. cooling rate (10^7 K sec^{-1})
2.	20.4	14.8	2.85 ± 0.75	0.255	45.4	1.88	0.84
3.	20.4	25.1	3.35 ± 0.70	0.165	32.0	1.66	1.20
5.	20.4	48.4	5.45 ± 0.50	0.150	24.7	1.30	1.55
33.	20.4	87.9	5.80 ± 0.65	0.145	16.7	0.93	2.29
46.	62.3	13.0	3.40 ± 0.70	0.600	73.0	3.23	0.57
67.	62.3	62.8	5.95 ± 0.80	0.270	22.1	1.39	1.89
10.	97.0	21.8	3.95 ± 1.10	0.380	60.8	2.65	1.20
28.	97.0	62.8	4.70 ± 0.70	0.200	20.1	1.17	3.63
30.	97.0	95.9	5.70 ± 0.55	0.125	15.0	0.98	4.82
15.	269.0	30.8	2.80 ± 0.55	0.360	46.6	2.77	2.06
29.	269.0	74.8	4.20 ± 0.55	0.125	20.7	1.48	4.62

depends upon the amplitude, brightness, contrast and magnification settings. However, the experimental procedure used to quantify the roughness condition of the centrifuge melt-spun ribbons yields representative roughness parameter values. These values are related to the extraction velocity, and depend upon the practiced ejection pressure. Work is now in progress to determine the optimal amplitude, brightness and contrast conditions to be used for SEM Y-modulation and scanning, as well as to elaborate a calibration procedure. The main advantage of the present technique is that it enables the scanning of the entire width of the ribbon. Scanning is fast and easy. While the digitizing procedure is lengthy, it affords permanent recording of the profiles in computer disks.

Contrary to what was expected, namely that lower DAS values should reveal smoother rim-substrate wetting pattern, it seems that increased roughness of the rim-contact side of the solidifying ribbons does not impede heat transfer. Despite the additional roughness resulting from increasing substrate velocities, the heat transfer ability of the melt to the substrate is improved, both by high extraction velocities and by high ejection pressures. It has been reported in previous work [4], that there is an increase in heat transfer coefficient when a matte surface is used, instead of a smooth one, in conventional melt-spinning. In CMS however, it appears that substantial mechanical dragging does occur, consecutive to the counter-rotating of the crucible and the rim. Such a mechanical dragging causes the ribbon to achieve thin section much faster than in conventional melt-spinning, thus enabling heat extraction in a more efficient way, despite the "bad" thermal contact conditions.

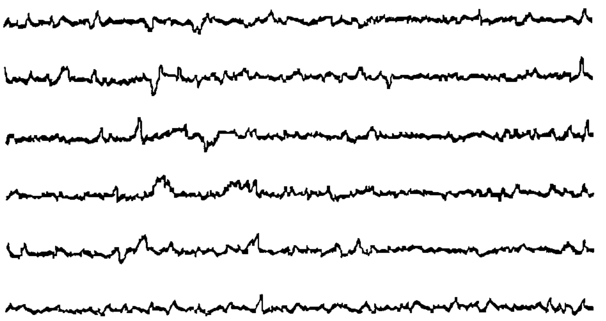

Figure 5 Digitized profiles on the computer screen.

The dragging mechanism consists in shear extension during cooling due to forces established as a result of the differences between the liquid ejection velocity and the extraction velocity, i.e. a drawing mechanism. The maximum theoretical amount of drawing is governed by the material continuity equation. The final drawn-down thickness should therefore be given by (if dragging is the only operative mechanism):

$$th_f = d(V_l/V_{tot})$$

where d is the diameter of the liquid jet, i.e. the crucible's orifice diameter (1 mm in our case), V_l is the ejection velocity of the liquid melt, V_{tot} is the extraction velocity (which is the sum of the crucible and substrate velocities).

The ejection velocity of the liquid melt depends upon the rotating velocity of the crucible. The actual velocities have been calculated according to the hydraulic model for CMS [5]. They range from 6.75 m sec^{-1} for the 1.8 kPa ejection pressure up to 26.28 m sec^{-1} for 269 kPa. The "drawing coefficient" V_l/V_{tot} is higher at low ejection pressures (for identical substrate velocities) and increases as substrate velocities increase (when ejection pressure remains constant). It is obvious that mechanical dragging is not the unique

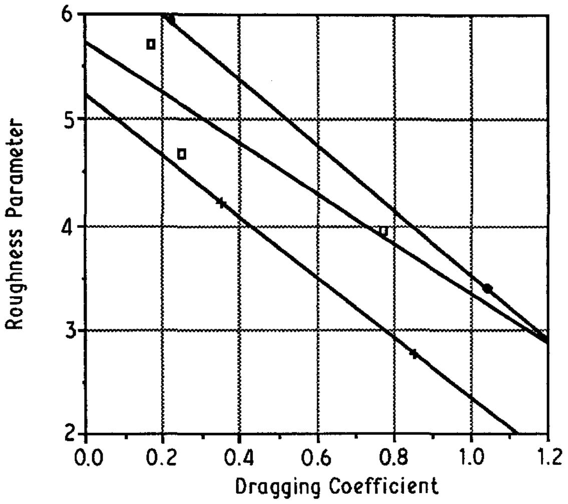

Figure 6 Roughness parameter plotted against dragging coefficient. (●) P_{ej} = 62.3 kPa, (□) P_{ej} = 97 kPa, (+) P_{ej} = 269 kPa.

mechanism influencing the ribbon thickness in CMS. But its contribution to the roughness parameter values is consistent with the experimental results. Fig. 6 shows how the roughness parameter values indeed decrease as the "dragging coefficient" increases and how they are undoubtedly lowered as the ejection pressure increases.

4. Conclusions

The wetting pattern characterization of rapidly solidified melt-spun metallic Al–Ge ribbons has been enabled by measuring the roughness of the rim-contact side of the ribbons. Roughness parameter values were obtained by combining SEM examination of the smooth surfaces, using amplitude modulation and line scanning, and micro-computerized digitizing of the line profiles. In such a manner, quantification of the wetting patterns is possible. As a result, the respective contribution of the ejection pressure and the extraction velocity on the wetting patterns of the solidifying ribbons is estimated. In CMS, the roughness of the rim-contact surface of the ribbons increases when extraction velocities are increased. Despite the additional roughness and the "bad" thermal contact conditions for effective heat transfer, high cooling rates (as measured by DAS), are achieved. It appears that a kind of dragging occurs, reducing the ribbon thickness by a shear drawing mechanism. The behaviour is characteristic of CMS, due to the specific hydraulic features involved with the counter-rotating of the crucible and the substrate. The exact nature of the liquid alloy stream and the heat transfer process, as well as their beneficial impact on the alloy solidification kinetics is presented elsewhere [5].

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