Fatigue of metallic glasses

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The fatigue behaviour of Ni₄₉Fe₂₉P₁₄B₆Si₂, Ni₄₈Fe₂₉P₁₄B₆Al₃ and Pd_{77.5}Cu₆Si_{16.5} metallic glasses is examined. In the finite lifetime regime the relationship between stress amplitude (σ_a) , fracture stress (σ_f) , mean stress (σ_m) and cycles to failure (N_f) is $\sigma_a = A(\sigma_f - \sigma_m)$ $(2N_f)^b$, where A and b are 16.9 and -0.40 respectively for reduced gauge section Ni₄₉ strips (for $\sigma_m \leq 140 \text{ kg mm}^{-2}$) and 27.0 and -0.44 for Pd base wires. These results are unusual in that $A \gg 1$. Consequently, a sharp discontinuity exists near $\sigma_a/(\sigma_f - \sigma_m) \simeq 1$. In a simple tensile test failure occurs at $\sigma_f(=\sigma_y)$ and $2N_f = 1$; for peak stresses only a percent or so less than σ_f the sample will withstand hundreds of cycles of stress. For uniform cross-section glassy metal filaments, a fatigue limit is observed at stress ratios (σ_a/σ_f) in the vicinity of 0.07 to 0.15. The fatigue limit for reduced section specimens is a factor of ~ 2 higher. Fatigue failure of the Ni–Fe strips may occur under partially or fully plane stress or plane strain conditions, depending on sample thickness and stress. Final failure of the Pd_{77.5}Cu₆Si_{16.5} wires always occurs by general yielding of the remaining section.

1. Introduction

Glassy metallic alloys are typically found near the composition $M_{80}X_{20}$ (in at. %), where M is a transition metal and X is one or more of the metalloids B, C, Si, P, etc, Pd₈₀Si₂₀ being one of the earliest examples [1]. Of more recent discovery are the glassy alloys of the approximate formula $(Ni_z Fe_{1-z})_{80} P_{14} B_6 [2, 3]$. These latter alloys exhibit tensile strengths of the order of 240 kg mm^{-2} $(340 \times 10^3 \text{ psi})$ [4, 5]. In addition, it has been demonstrated that such alloys may be made in continuous lengths, with "finished" cross-sectional dimensions, in a single step, i.e. by continuous rapid quenching from the melt [6]. This favourable combination of parameters, i.e. high strength, reasonable cost of constituent elements and economy of fabrication, suggests the use of these glassy metal filaments for structural reinforcement applications, It, therefore, becomes of interest to further characterize the mechanics of these materials, particularly with respect to their fracture toughness and response to cyclic loading.

A few reports on these subjects have appeared recently. The mode III fracture toughnesses of a

The purpose of the present paper is to further consider the fatigue behaviour of glassy alloys. Results are presented for two closely related Ni–Fe base METGLAS[®]* alloys, Ni₄₉Fe₂₉P₁₄B₆Si₂ and Ni₄₈Fe₂₉P₁₄B₆Al₃. Results are also presented for glassy Pd_{77,5}Cu₆Si_{16,5}.

2. Experimental procedure

2.1. Materials

The alloys studied were quenched directly from the melt and adjudged to be glassy by X-ray examination. The Ni₄₈ alloy was prepared in strip form. Sections $\sim 7 \text{ cm}$ long were cut and polished on their narrow edges to produce samples 0.4 mm wide by $\sim 0.026 \text{ mm}$ thick. Strips of the Ni₄₉ alloy with lightly polished edges were $\sim 0.76 \text{ mm}$ wide by $\sim 0.076 \text{ mm}$ thick. The Pd_{77.5} Cu₆Si_{16.5} was in

few metallic glasses were reported by Kimura and Masumoto [7]. The fatigue behaviour of $Pd_{80}Si_{20}$ strips has been reported by Ogura *et al.* [8]. The author has examined the dependence of fracture toughness on sample thickness for Ni–Fe base alloys [9] and determined the fatigue crack propagation rate for a Ni₃₉Fe₃₈P₁₄B₆Al₃ alloy [9].

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wire form and had a diameter of ~ 0.28 mm.

As produced, the Pd alloy and Ni₄₉ alloy [5] exhibit tensile yield strengths of ~ 147 ± 3 and ~ 243 ± 7 kg mm⁻², respectively. In the latter case this value was determined on hand polished, reduced gauge section specimens. The small dimensions of the Ni₄₈ strips precluded preparation of such specimens. It has been observed, however, that the ratio of yield stress to Vicker's hardness (σ_y/H) is constant at ~ 3.2 for the Pd and Ni₄₉ alloys [11]. As the hardnesses of the Ni₄₉ (H = 792 kg mm⁻²) and Ni₄₈ (H = 752 kg mm⁻²) alloys are equivalent, within experimental error, we conclude that they have approximately the same yield strengths.

2.2. Specimen configuration

The Ni₄₈ specimens were tested as uniform strips with dimensions as noted above. Samples of the Ni₄₉ alloy were tested in "centre hole" and "gauged" configurations. In the former case a hole of 0.1 mm diameter was placed in the centre of a specimen by electric discharge machining (EDM). In the latter case a gauge section ~ 0.25 mm wide by ~ 1 mm long was hand polished on the specimen. Specimens of the Pd_{77.5}Cu₆Si_{16.5} amorphous alloy were tested as uniform cross-section filaments and, in a few cases, with an electropolished gauge section ($\simeq 0.18$ mm diameter by 6 mm).

2.3. Mechanical testing equipment

The majority of the experiments were conducted using an MTS electro-hydraulic, servo-controlled mechanical testing machine [12]. Cyclic loading with a sinusoidal wave form, was conducted in load control at a frequency on the order of 0.5 to 5 Hz. The load signal was monitored by an amplitude measurement panel [12] and displayed on an oscilloscope.

For a number of long term tests specimens were cyclically loaded in 22.7 kg (50 lb) capacity Sonntag fatigue machines [13]. In these machines the mean stress is provided by spring loading and the oscillating stress is provided by an eccentric weight rotating at 30 Hz; they were calibrated using an MTS load cell monitored by MTS electronic conditioning units.

All tests were conducted in the tension-tension mode. In order to eliminate slack in the loading system the minimum stress applied was $\gtrsim 3.5$ kg mm⁻² (5 × 10³ psi). Drum-clamp type grips designed as described previously [9], were used for

many specimens. It was found more convenient to use simple flat clamp grips for the reduced gauge section specimens.

All tests were conducted at room temperature ($\sim 22^{\circ}$ C) in the laboratory environment.

3. Results

3.1. Lifetime data

The sample lifetime data for the Ni-Fe alloys and the Pd_{77.5}Cu₆Si_{16.5} alloy are presented in Figs. 1 and 2, respectively. In each case the abscissa is the number of stress reversals to failure, i.e. twice the number of cycles to failure, $N_{\rm f}$. A simple uniaxial tensile test is considered to exhibit one reversal to failure. The ordinate is plotted as the stress ratio $S = \sigma_{\rm a}/(\sigma_{\rm y} - \sigma_{\rm m})$ where $\sigma_{\rm m}$ is the mean stress acting on the sample, $\sigma_{\rm y}$ is the yield strength of the material (equal to the fracture strength for properly prepared glassy alloy specimens) [5] and $\sigma_{\rm a}$ is the half-amplitude of the sinusoidal stress (load) programme, i.e. $\sigma_{\rm a} = (\sigma_{\rm max} - \sigma_{\rm min})/2$. For convenience $\sigma_{\rm a}$ is simply called the stress amplitude. Clearly, S cannot exceed 1.

For the Ni–Fe alloys the slight compositional difference is considered to be inconsequential [9]. Hence the observed variation in sample lifetime at constant S arises for a variety of other reasons. For example, the open circles pertain to uniform cross-section (Ni₄₈) strips, wherein failure is usually initiated at one of the contact points with the grips. The solid circles obtain for specimens (Ni₄₉) with a reduced cross-section gauge length. Elimination of the elastic constraint and/or the fretting action of the grips leads to longer life. The solid triangles pertain to (Ni₄₉) samples with an EDM hole. The severe stress concentration so introduced ($K_t = 3$) leads to reduced fatigue life.

The solid line at the top of the figure approximates the finite lifetime behaviour of gauged Ni_{49} specimens for mean stresses up to about 140 kg mm⁻² (which covers the zero-tension regime). For higher mean stress (169 kg mm⁻²) the data fall approximately parallel to, but above, the line.

The open symbols in Fig. 2 represent data for uniform cross-section $Pd_{77,5}Cu_6Si_{16,5}$ filaments tested under various constant mean stress or constant minimum stress conditions. The dashed line, shown for comparison, approximates the data of Ogura *et al.* [8] for $Pd_{80}Si_{20}$ strips (σ_y is taken as that of $Pd_{77,5}Cu_6Si_{16,5}$ because these alloys exhibit identical hardnesses [11, 14]). The solid circles pertain to specimens with a reduced cross-section



Figure 1 Stress versus reversals to failure $(2N_f)$ for Ni₄₈ Fe₂₉P₁₄B₆Al₃ and Ni₄₉Fe₂₉P₁₄B₆Si₂ metallic glasses. Stress is given in terms of the parameter $S = \sigma_a/(\sigma_y - \sigma_m)$ where σ_a = stress amplitude, σ_m = mean stress and σ_y = yield stress. The line given by $S = (2N_f)^{-0.06}$ is the upper limit of fatigue lifetimes observed for high strength steels [16]. Arrows indicate tests terminated without failure at the lifetime shown, except for S = 0.34, where N_f reached 1.2×10^8 .



Figure 2 Stress-lifetime behaviour for $Pd_{77.5}Cu_6Si_{16.5}$ metallic glass wires tested under various conditions of constant mean stress or constant minimum stress. Parameters are as in Fig. 1.



Figure 3 Combinations of stress amplitude (σ_a) and mean stress (σ_m) for lifetime $N_f \simeq 10^6$ for Ni₄₈ strips (triangles), Ni₄₉ gauged specimens (squares) and Pd_{77.5}Cu₈Si_{16.5} wires (circles).

gauge length.

For each type of specimen tested a fatigue limit (abrupt transition to long life as S is reduced) was observed. Combinations of σ_a and σ_m for $N_{\rm f} \gtrsim 10^6$ are shown in Fig. 3. For constant S the data should fall on a sloping straight line intersecting the abscissa (σ_m) at σ_f , as shown, for example, by the line $S \simeq 0.42$ (plotted for $\sigma_{\rm f} \simeq 250 \, \rm kg \, mm^{-2}$). For glassy alloys $S \neq \rm constant$; σ_a is relatively insensitive to σ_m in the regime examined and at high σ_m the curve must fall sharply to intersect $\sigma_m = \sigma_f$. For the uniform cross-section $Pd_{77,5}Cu_6Si_{16,5}$ specimens σ_a is approximately constant at 23 kg mm^{-2} (32.5×10^3 psi), i.e. the stress ratio for long life $(\sigma_a/\sigma_f) \simeq 0.15$. Long life is achieved at higher amplitude [41 kg mm⁻² $(59 \times 10^3 \text{ psi})$] only for the electropolished gauge specimen (Fig. 2, solid circle). For the reduced section Ni₄₉ strips σ_a decreases slowly with increasing $\sigma_{\rm m}$; $\sigma_{\rm a}/\sigma_{\rm f}$ is of the order of 0.13 to 0.17. For the Ni₄₈ strips $\sigma_a/\sigma_f \simeq 0.07$.

3.2. Fracture topology

The fatigue fracture topology of a Ni₄₉Fe₂₉P₁₄ B₆Si₂ specimen, with a reduced cross-section gauge length, is shown in Fig. 4. Initiation of the fatigue crack occurred near point A (crack initiation typically occurs at or near one of the narrow edges of the specimen). The fracture surface in the vicinity of the stably propagating fatigue crack is marked by periodic striations. According to measurements on a similar alloy (Ni₃₉Fe₃₈P₁₄B₆Al₃) the striation spacing is ~ 6 da/dn, where da/dn is the crack advance per cycle [9]. For the Ni₃₉ alloy da/dn (mm/cycle) ~ 2 × 10⁻⁸ $\Delta K^{2.25}$ where the cyclic



Figure 4 Fatigue fracture surface for a reduced gauge section $Ni_{49}Fe_{29}P_{14}B_6Si_2$ strip $[\sigma_a = 72 \text{ kg mm}^{-2} (102 \times 10^3 \text{ psi}); \sigma_m \equiv 79 \text{ kg mm}^{-2} (112 \times 10^3 \text{ psi})]$. The fatigue crack was initiated near point A. Unstable crack propagation initiated along the curved front marked by the arrows. The crack moved from left to right.

stress intensity, ΔK , has units of kg mm^{-3/2}.

Unstable crack propagation begins along a semicircular front marked by the arrows. For the most part, the crack propagates in the direction of the width vector (left to right) producing a classical chevron pattern. On a gross scale the chevron pattern lies at 90° to the tensile axis, i.e. square fracture occurs. On a finer scale, however, the chevrons exhibit a saw-tooth morphology marked by a fine scale equi-axed vein pattern, indicating that local failure occurs by shear rupture (see [9]). Shear lips lie at the edges of the specimen parallel to the direction of crack propagation. For the EDM hole specimens the fracture features on either side of the hole are identical to those in Fig. 4.

As has been reported previously [9] the Ni₄₉ specimens are sufficiently strong and thick so that crack propagation occurs under fully plane conditions ($K_{IC} \simeq 30 \text{ kg mm}^{-3/2}$). A transition from square (plane strain) to slant (plane stress) fracture typically occurs during crack propagation in the thinner Ni₄₈ specimens [9]. For very high cyclic stress (σ_{max} of the order 0.9 σ_y) the square to slant transition also occurs for Ni₄₉ specimens. In one case ($\sigma_{max} \simeq 0.99 \sigma_y$) failure occurred in the 53° mode (general yielding) [5].

The fracture surface of a reduced gauge section $Pd_{77,5}Cu_6Si_{16,5}$ filament is shown in Fig. 5 (the fracture topology is identical for uniform filaments). Fatigue crack initiation occurred at a site diametrically opposite to the site marked B. Surface striations are evident in the mottled, stable fatigue crack region (Fig. 6a and b). The catastrophic fracture surface is marked by an equi-axed vein pattern typical of shear rupture. The appear-



Figure 5 Fatigue fracture surface of a reduced gauge section $Pd_{77.5}Cu_6Si_{16.5}$ filament. The fatigue crack initiated to the left of the surface (see Fig. 6 for detail). The remainder of the section is marked by a smooth shear offset (near B) and vein pattern typical of shear rupture. This indicates that final failure occurs by yielding of the remaining section. On a macroscopic scale the fracture surface slopes down from left to right; the normal to this surface lies at ~ 45° to the wire axis (see [10]).

ance of this surface is identical to that observed in simple tensile failure [10, 15] which occurs coincident with yielding [10]. Hence it is apparent that fatigue failure in $Pd_{77.5}Cu_6Si_{16.5}$ filaments occurs only when the load bearing section is sufficiently reduced to allow general yielding of the specimen under the applied peak load.



Figure 6 (a) An enlarged view of the fatigue crack in Fig. 5. (Micrograph rotated 90° anti-clockwise relative to Fig. 5.) (b) An enlarged view, centered about the black dot in (a) showing the surface striations created by stable fatigue crack propagation.

4. Discussion

4.1. Finite lifetime regime

The solid lines (finite lifetime data) shown in Figs. 1 and 2 may be approximated by a formula of the type

$$\sigma_{\mathbf{a}} = A(\sigma_{\mathbf{f}} - \sigma_{\mathbf{m}})(2N_{\mathbf{f}})^{b}$$
(1)

where the true fracture stress $\sigma_{\rm f} = \sigma_{\rm y}$ and the other symbols are as defined above. For Pd_{77.5} Cu₆Si_{16.5} the values of A and b are 27 and -0.44 respectively; these parameters appear to hold for all mean stresses. For the Ni₄₉Fe₂₉P₁₄B₆Si₂ specimens $A \simeq 16.9$ and $b \simeq -0.40$ for mean stresses less than ~ 140 kg mm⁻²; for higher mean stress the line appears to be shifted parallel to the right, i.e. A increases. Within the range of the data, the

parameters given yield lines that are practically coincident.

The form of Equation 1 is the same as that observed for the stress amplitude-lifetime behaviour of crystalline metals, with the significant difference that, in the latter case, $S = \sigma_a/(\sigma_f - \sigma_m) \simeq 1$ at $2N_f = 1$, i.e. A = 1 (see e.g. the review by Landgraf [16]). In effect the fatigue behaviour of metallic glasses at high cyclic stresses, i.e. high S, is biomodal. In a simple tensile test the sample fails on the first excursion to $\sigma_{max} = \sigma_f$; with only slight reduction of the cyclic peak stress it will sustain 600 or more stress cycles. This was examined most closely for the Ni₄₉ specimens where repeated stresses (σ_{max}) within $\sim 1\%$ of σ_f can be sustained. No lifetimes between $N_f = 1/2$ and 600 were observed. In the finite lifetime regime $2N_{\rm f}$ increases only slightly with decreasing S, i.e. |b| is somewhat larger than typically observed for crystalline metals (~ 0.06 to 0.10). As values of S in excess of 1 are not physically realizable, Equation 1 (with A > 1) must, obviously, be considered as an interpolation equation.

It is important to emphasize that the lifetimes observed for the Ni₄₉ specimens at high S are about three orders of magnitude greater than may be expected for high strength steels $[S \simeq (2N_f)^{0.06}]$. Indeed this long life is observed in a high tensile mean stress regime, under load control, where a crystalline material would typically fail by excessive creep. This would occur because, while the true fracture stress of a material such as 250 maraging steel is $\sim 250 \,\mathrm{kg}\,\mathrm{mm}^{-2}$ ($355 \times 10^3 \,\mathrm{psi}$), its yield strength is only about $180 \,\mathrm{kg}\,\mathrm{mm}^{-2}$ ($260 \times 10^3 \,\mathrm{psi}$) [17]. It appears that the unusual bimodal fatigue behaviour of properly prepared metallic glass specimens, such as the gauged Ni₄₉ specimens, is a direct result of the coincidence of yielding and fracture.

From previous work [9] it is known that the fatigue crack propagation rate in Ni–Fe base glassy alloys is similar to that in high strength steels. It is also known that $K_{\rm IC}$ (~ 30 kg mm⁻²) is lower than that of, for example, maraging steel. Hence we conclude that the long lifetimes at high S exhibited by glassy metals must result due to a difficulty of crack initiation. In essence these materials behave as elastic-perfectly plastic solids, i.e. elastic behaviour is retained at stresses up to the yield and fracture stress. Hence it is difficult to generate the cyclic plastic strain required for crack initiation [18].

The fatigue lifetime at high S, e.g. the $2N_{\rm f}$ intercept at S = 1, will depend on the nature and distribution of stress concentrating defects, such as inclusions or voids, in the material. It will also depend on testing conditions. In ungauged specimens fretting with the grips will lead to premature crack initiation, shifting the lifetime to lower values. Placing a hole in the material has the same effect.

In the case of $Pd_{77.5}Cu_6Si_{16.5}$ wires, fretting with the grips also occurs. In this case, however, the $2N_f$ intercept at S = 1 is still $> 10^3$. This probably occurs, at least in part, because the final failure mode (general yielding) is a high toughness mode. If one avoids extrinsic effects, e.g. fretting, fatigue lifetimes should approach extreme values for all stresses up to, but excluding, the yield stress as the glassy metal is produced in an increasingly cleaner form.

4.2. Long life regime

A subject of technological interest is the value of S or σ_a/σ_f (when the mean stress is relatively unimportant) for which long life $(N_f > 10^6 \text{ to } 10^7)$ is observed. This depends, of course, on the sample and/or test conditions, e.g. the long life limit (σ_a/σ_f) for gauged specimens of Ni₄₉Fe₂₉P₁₄B₆Si₂ and Pd77.5Cu6Si16.5 is a factor of about two greater than for uniform cross-section specimens. The lower value is important because it applies to a configuration of practical significance, i.e. concentrated stresses and/or fretting will often occur in, e.g. filament reinforcement applications. For thin strips of the Ni–Fe alloy σ_a/σ_v equals ~0.07; $\sigma_{\rm a} \simeq 17.6 \, \rm kg \, mm^{-2} (25 \times 10^3 \, \rm psi)$ for $\sigma_{\rm min} = 21 \, \rm kg$ mm⁻² and is insensitive to σ_m (Fig. 3). Hence for zero-tension loading, long life would obtain for $\sigma_{\rm a} = \sigma_{\rm m} \simeq 18 \, \rm kg \, mm^{-2}$ and the conventional endurance limit $\sigma_{\rm L}$ ($\sigma_{\rm a}$ for $\sigma_{\rm m} = 0$) would be ~ 18 kg mm^{-2} .

This value may be compared with $\sigma_{\rm L} \simeq 35$ kg mm⁻² (50 × 10³ psi) for severely notched ausformed H-11 steel [19]. This steel has a tensile strength of 250 kg mm⁻² (355 × 10³ psi) and, when unnotched, apparently exhibits the highest endurance limit (118 kg mm⁻²; 168 × 10³ psi) known for steels.

A comparison also of interest derives from measurements of the fatigue behaviour of brasscoated steel monifilaments (0.25 mm diameter) from a commercial tire cord. These filaments exhibit a tensile strength of ~ 265 kg mm⁻². They were tested as part of this study under conditions identical to those used for the glassy metals. For these samples the long life regime is reached at $\sigma_a \simeq 32 \text{ kg mm}^{-2}$ ($45 \times 10^3 \text{ psi}$) with $\sigma_{\min} \simeq 21$ kg mm⁻². This value is again comparable to that observed for the Ni–Fe glassy strips.

For the unnotched (and unpolished) Ni₄₉ strips $\sigma_{\rm L}$, as deduced from Fig. 3, is about 50 kg mm⁻², which is about one half that for maraging steel [16] (shown as the line $S \simeq 0.42$). The long life behaviour for the gauged Ni₄₉ strips approaches that for maraging steel as $\sigma_{\rm m}$ increases.

The $Pd_{77.5}Cu_6Si_{16.5}$ wires exhibit a ratio $\sigma_a/\sigma_f \simeq 0.15$ for $N_f \ge 10^6$ (as compared to ~ 0.07 for uniform section NiFe strips). We expect that this higher value may result because of geometry. That is, in uniaxial tension or cyclic loading high aspect

ratio (\gtrsim 7) strips fail by mode III (tearing) and/or mode I (plane strain) processes. These are lower energy modes of failure than yielding, the failure mode of the Pd alloy wire; also crack initiation is probably easier in a strip than in a wire. Using the 0.15 ratio one may expect a fatigue limit of $\sigma_a \simeq 36 \text{ kg mm}^{-2}$ ($\sigma_f \simeq 243 \text{ kg mm}^{-2}$) for Ni-Fe wires or very low (\ll 7) aspect ratio strips.

5. Conclusion

The finite lifetime relationship between stress and cycles to failure for metallic glasses is of the form $\sigma_a = A(\sigma_f - \sigma_m) (2N_f)^b$ where A and b are 16.9 and -0.40 respectively for Ni₄₉Fe₂₉P₁₄B₆Si₂ (for $\sigma_{\rm m} \lesssim 140 \, \rm kg \, mm^{-2}$) and 27.0 and -0.44 for Pd_{77} ₅Cu₆Si₁₆ ₅. This relation is of the same form as for crystalline metals with the distinct exception that, in the latter case, $A \simeq 1$. Therefore, for large values of the ratio $S = \sigma_a/(\sigma_f - \sigma_m)$ metallic glasses exhibit lifetimes about three orders of magnitude greater than, for example, a steel of comparable fracture strength. For the Ni49 metallic glass, where it was studied most closely, a sharp discontinuity exists near $S \simeq 1$; the sample exhibits a lifetime of >600 cycles for cyclic peak stresses with a percent or so of σ_{f} .

A long life limit $(N_f \gtrsim 10^6)$ is reached for stress ratios (σ_a/σ_y) of ~0.07 and ~0.15 for uniform cross-section filaments of the Ni-Fe and Pd base glasses respectively. For reduced section specimens the fatigue limit is a factor of ~2 higher. The lifetime is insensitive to mean stress in this regime. It is suggested that the higher σ_a/σ_y ratio observed for the Pd base wires may be geometrical effect, i.e. Ni₄₉ wires would exhibit a ratio of 0.15.

Failure occurs under partially or fully plane

strain crack propagation conditions in the Ni–Fe base strips. Final failure by general yielding occurs only for peak cyclic stresses within a percent or so of the yield stress. Failure of the $Pd_{77,5}Cu_6Si_{16.5}$ wires always occurs by general yielding.

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