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# WEAR AND CORROSION PERFORMANCE OF METALLURGICAL COATINGS IN SODIUM

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# Wear and Corrosion Performance of Metallurgical Coatings in Sodium

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## ABSTRACT

This paper reviews the friction, wear, and corrosion performance of several metallurgical coatings in 200°-650°C sodium. Emphasis is placed on those coatings which have successfully passed the qualification tests necessary for acceptance in breeder reactor environments. Tests include friction, wear, corrosion, thermal cycling, self-welding, and irradiation exposure under as-prototypic-as-possible service conditions. Materials tested were coatings of various refractory metal carbides in metallic binders, nickel-base and cobalt-base alloys and intermetallic compounds such as the aluminides and borides. Coating processes evaluated included plasma spray, detonation gun, sputtering, spark-deposition, and solid-state diffusion.

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## INTRODUCTION

Sodium is an exceptionally effective heat transfer fluid for high temperature systems, and has been chosen for use in certain breeder reactor concepts. For the past decade, the U.S. Department of Energy, and its predecessor agencies, have sponsored research in materials and coatings for use in these high temperature liquid metal systems.

Sodium provides a relatively benign environment for most structural materials. However, from a tribological (wear and friction) viewpoint, sodium presents a challenging environment. Sodium is a highly reactive element that tends to strip the oxide films from most metal surfaces, leaving them in an ultra-clean condition. This promotes adhesive wear, high friction, and self-welding tendencies that are similar to those observed in a high vacuum. In most other environments, lubricants could be used to reduce friction and to prevent galling. But sodium also reacts with nearly all known lubricants.

Under certain conditions, sodium can react with elements in a metal surface to create films of sodium-metal-oxygen complexes (such as $NaCrO_2$). These films are lubricative and can significantly reduce friction coefficients and galling tendencies. The effect of such films was illustrated during friction and wear testing of chromium carbide + nickel-chrome coatings in sodium. Friction coefficients varied from as high as 0.9 when no film was present, to less than 0.5 on the same surface at the same temperature after the film had been allowed to form(1). Similar effects have been noted on a wide variety of materials, including austenitic stainless steels, Inconel 718*, and Stellite 6**.

The formation and effectiveness of these films are functions of temperature, oxygen-content of the sodium, activity of the reacting elements in the rubbing surfaces, and the test or service conditions (i.e. interface geometries, rubbing velocities, loads, etc.). Unfortunately, the kinetics and stability of these film formations are subject to so many variables that the films generally cannot be relied upon for acceptable performance of mechanical components in sodium.

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*Trademark, International Nickel Co.
**Trademark, Cabot Corp.

When high friction or galling cannot be tolerated in a given component, the most effective solution has been to use a hard, wear-resistant metallurgical coating on the component structural material. The coatings that consistently exhibit the lowest friction coefficients and the least surface damage effects are those that are not only very hard and abrasion-resistant, but that are also capable of reacting with the sodium to form the complex sodium-oxygen-metal films (i.e. $NaCrO_2$ or $NaAlO_2$). This paper reviews the significant advances in the development and qualification of these coatings for tribological applications in breeder reactor environments.

## COATINGS QUALIFIED FOR USE IN SODIUM COOLED REACTORS

Typical breeder reactor service conditions involve sodium exposures of one to 40 years, neutron irradiation to as high as $10^{23}$ neutrons/$cm^2$, and temperatures to 600°C or more. Qualification of coatings has required extensive testing of a large variety of materials and coating processes to identify those capable of providing adequate performance under these demanding conditions. Qualification tests include friction, wear, corrosion, self-welding, thermal cycling, and irradiation tests under as-prototypic-as-possible service conditions.

Of the materials and processes tested, only a detonation gun (D-gun) applied chromium carbide in nichrome (Ni-Cr) or Inconel 718 binder, and a nickel-aluminide diffusion coating on Inconel 718 have successfully passed all qualification tests. A coating of 100% Tribaloy 700* (a nickel-base hardfacing alloy) and a coating of chromium carbide plus 15 vol. % Tribaloy 700, both applied by detonation-gun to Type 316 stainless steel, are in the final phase of testing. All other coating processes and materials tested have failed one or more of the qualification tests, or are still in qualification testing.

Other coating processes tested include plasma spray, spark deposition, sputtering, boriding and chromizing. Most of the plasma spray coatings failed the thermal cycling tests and all of them examined to date failed the irradiation tests. Sputtered coatings cracked and spalled in 625°C sodium and in the irradiation tests. Spark deposition coatings, although performing well in screening tests, were not tested further due to the process development work that would be required and the early success of the detonation gun coatings.

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*Trademark, Cabot Corp.

The spark deposition coatings, however, still hold promise and development efforts are continuing. Chromium diffusion (chromizing) and boride diffusion (boriding) coatings were tested on several austenitic stainless steels and nickel-base alloys. The chromized coatings produced some improvement in friction coefficients for most of the materials tested, but wear performance did not consistently improve. Although resistance to surface damage improved, in most cases galling was still reported. Some of the borided coatings spalled during testing in high temperature sodium due to formation of a low-melting nickel-boride phase in the coating interface. Further development of boride coatings was deferred because their use would be restricted to non-neutron environments.

The chromium carbides were identified in friction screening tests early in the program, as exhibiting the lowest and most consistent friction coefficients in 200° to 600°C sodium(2,3). Other bonded carbides, although frequently showing good wear-resistance, produced much higher friction coefficients and were dropped from the program.

## PROPERTIES OF DETONATION GUN APPLIED COATINGS

### Chromium Carbide Coating

Friction and wear tests were performed on a detonation gun applied chromium carbide + 15 volume percent nichrome binder on a Type 316 stainless steel substrate in 232° to 627°C sodium at 2 to 7 MPa loads(4). Dynamic friction results are shown in Figure 1. At all temperatures investigated, the chromium carbide coatings displayed dynamic friction coefficients considerably lower than those for uncoated Type 316 stainless steel. Friction coefficients tended to increase with cumulative rubbing distance and usually stayed within the 0.2 to 0.6 range all test temperatures up to nearly 600°C. At 627°C a significant rise in friction to the 0.8 to 0.9 range was observed.

Not only do the chromium carbide coatings produce a significant reduction in friction coefficients when compared to uncoated Type 316 stainless steel, but more significantly a different wear behavior is also observed. Austenitic stainless steels exhibit the classic adhesive wear behavior when rubbed against themselves in sodium, exhibiting at times severe surface damage. In contrast, applying a D-gun coating of chromium carbide to both surfaces and testing under the same conditions results in an abrasive wear behavior with a minimum of surface damage. No significant wear of D-gun chromium carbide coatings has been observed after nearly 210 m of cumulative rubbing in 232° to 627° sodium at contact

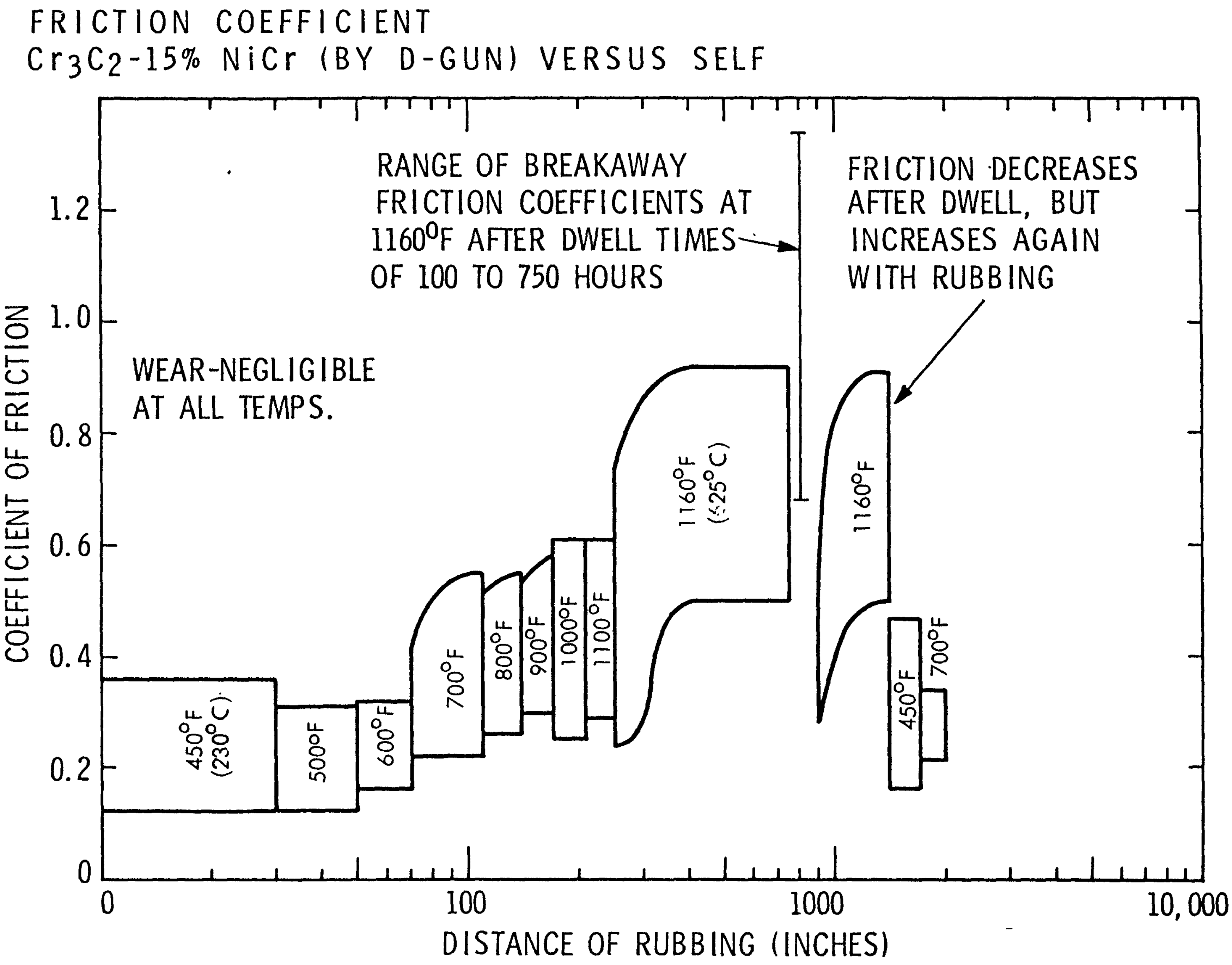

Figure 1 - Friction of Chromium Carbide + 15 vol.% Nichrome Detonation Gun Applied Coating in Sodium. (Ref. 1)

pressures of up to 7 MPa. Observed surface damage has been limited to a polishing and smoothing of high surface asperities with no significant material removal.

A significant reduction in self-welding of Type 316 stainless steel in high temperature sodium can be achieved by the application of a detonation gun chromium carbide coating to both rubbing surfaces. The range of self-welding coefficients expressed as $\Delta\mu$ (breakaway friction minus average dynamic friction coefficients) for coated and uncoated Type 316 stainless steel is shown in Figure 2. These data are from a series of dwell-breakaway friction tests of increasing dwell times from 1-100 minutes in 427° to 649°C sodium at varying contact pressures (2 to 20 MPa).

Sodium compatability testing in 625°C flowing sodium for over 18,000 hours of exposure has revealed corrosion rates of just over 2.5 μm/yr., well within the qualification requirement. The primary effect of sodium exposure appears to be selective removal of the nichrome binder, but without apparent loss of coating integrity. A comparison of the corrosion behavior of four low friction, wear resistant coatings for sodium service is shown in Figure 3.

Coating stability was evaluated under the following thermal cycle conditions:

- Hold at 627°C for from 8 hr to 4 days
- Quench in < 1 minute to 427°C in sodium
- Raise temperature of sodium at rate of 28° C/h to 627°C
- Repeat cycle

D-gun applied chromium carbide coatings have withstood 120 such cycles without any detectable deterioration.

Coatings irradiated to $1.2 \times 10^{22}$ n/cm$^2$ (E > 0.1 MeV) have shown no detectable deterioration such as cracking, spalling, or flaking. Dynamic friction test results indicate friction coefficients equal to or lower than unirradiated coatings particularly at 627°C where irradiated coatings exhibited dynamic friction coefficients of from 0.4 to 0.65, compared to 0.35 to 0.85 for unirradiated coatings.

### Tribaloy 700 Coatings

Tribaloy 700 is a nickel-base laves hardened hardsurfacing alloy of composition 50 wt % Ni, 32 wt % Mo, 15 wt % Cr, and 3 wt % Si. Detonation gun coatings of 100% Tribaloy 700 have exhibited among the lowest friction coefficients

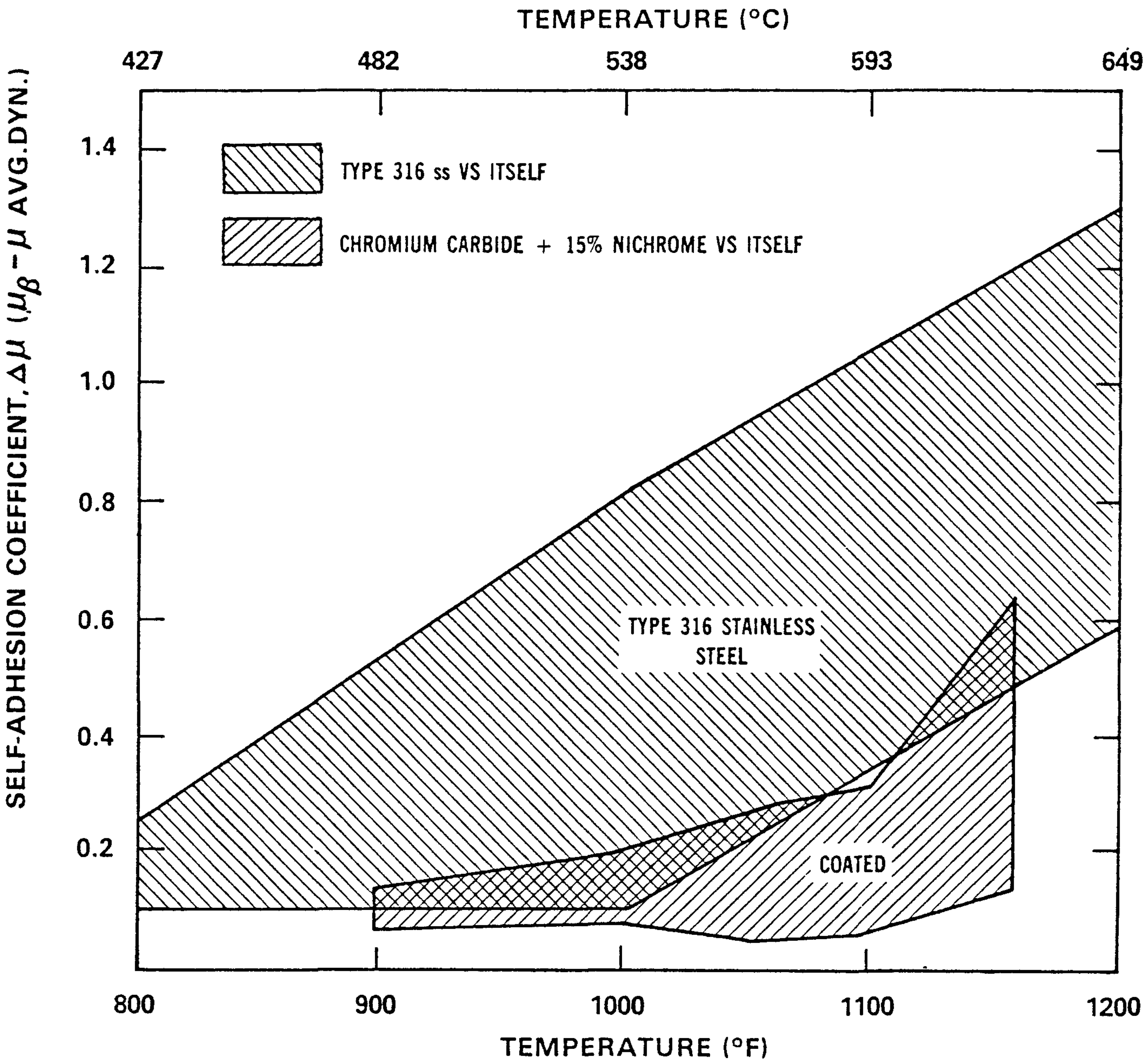

Figure 2 - Effect of Chromium Carbide Detonation Gun Coating on the Self-Welding of Type 316 Stainless Steel in Sodium. (Ref. 4)

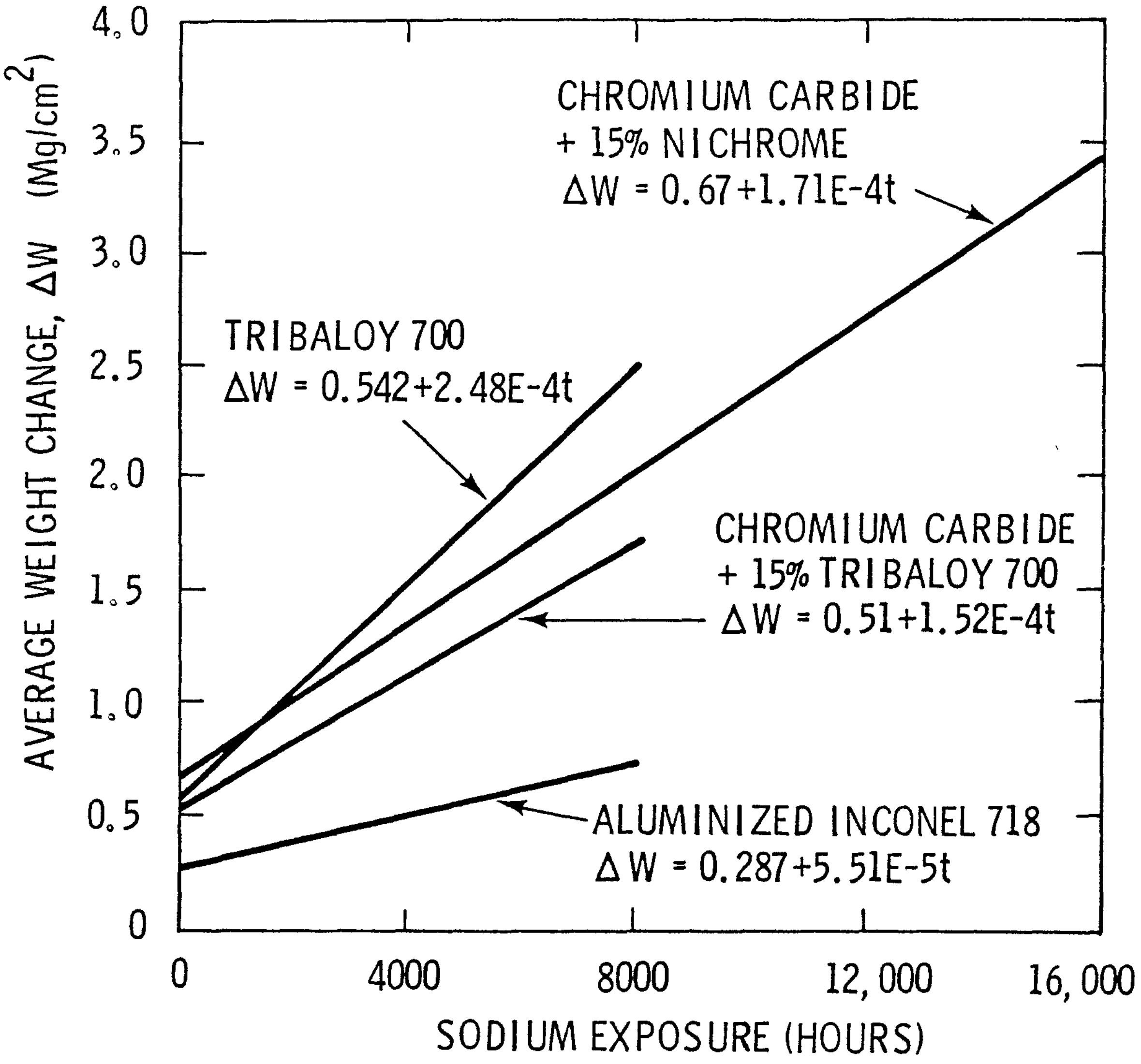

Figure 3 - Corrosion of Four Wear Resistant Coatings in 627° C Sodium.

measured for metallic materials in high temperature sodium(5). Friction and wear tests conducted in 200° to 649°C sodium indicate that Tribaloy 700 coatings exhibit up to a 50% reduction in dynamic friction coefficients when compared to chromium carbide + nichrome coatings under identical test conditions as shown in Figure 4. The wear behavior of Tribaloy 700 is characterized as a mild abrasive wear comparable to the chromium-carbide + nichrome coating. Unfortunately, the Tribaloy 700 seems to show a greater self-welding tendency than the chromium-carbide + nichrome coatings, particularly in > 600°C sodium. In addition Tribaloy 700 coatings exhibited corrosion rates similar to those exhibited by the chromium-carbide + nichrome coatings, as shown in Figure 3.

The coatings have passed thermal cycling tests (120 cycles) without any detectable deterioration. Irradiation testing of the Tribaloy 700 coatings is now in progress. The Tribaloy 700 coatings will be considered qualified for reactor-core use upon successful completion of these tests.

Chromium Carbide and Tribaloy 700 Coating

In an attempt to improve the high temperature (> 600°C) sodium friction behavior and self-welding tendency of the nichrome binder series of chromium carbide coatings, various metallic binders were tried. The most successful attempt to date appears to be the substitution of Tribaloy 700 for the nichrome in the reference chromium carbide coating.

Dynamic friction testing of D-gun applied chromium carbide + 15 vol. % Tribaloy 700 on Type 316 stainless steel in 627°C sodium have shown significant improvement in the friction behavior over the chrome carbide + nichrome coatings(4). Also breakaway friction test results indicate a marked improvement in the self-welding tendencies of this coating over the nichrome binder coatings, in 627°C sodium.

Sodium compatability tests in 627°C flowing sodium for exposure times of up to 8000 hours have revealed corrosion rates for this coating equal to or lower than those of the nicrhome binder coating. (See Figure 3.)

This coating has passed thermal cycling tests (120 cycles), and is now in long term irradiation testing.

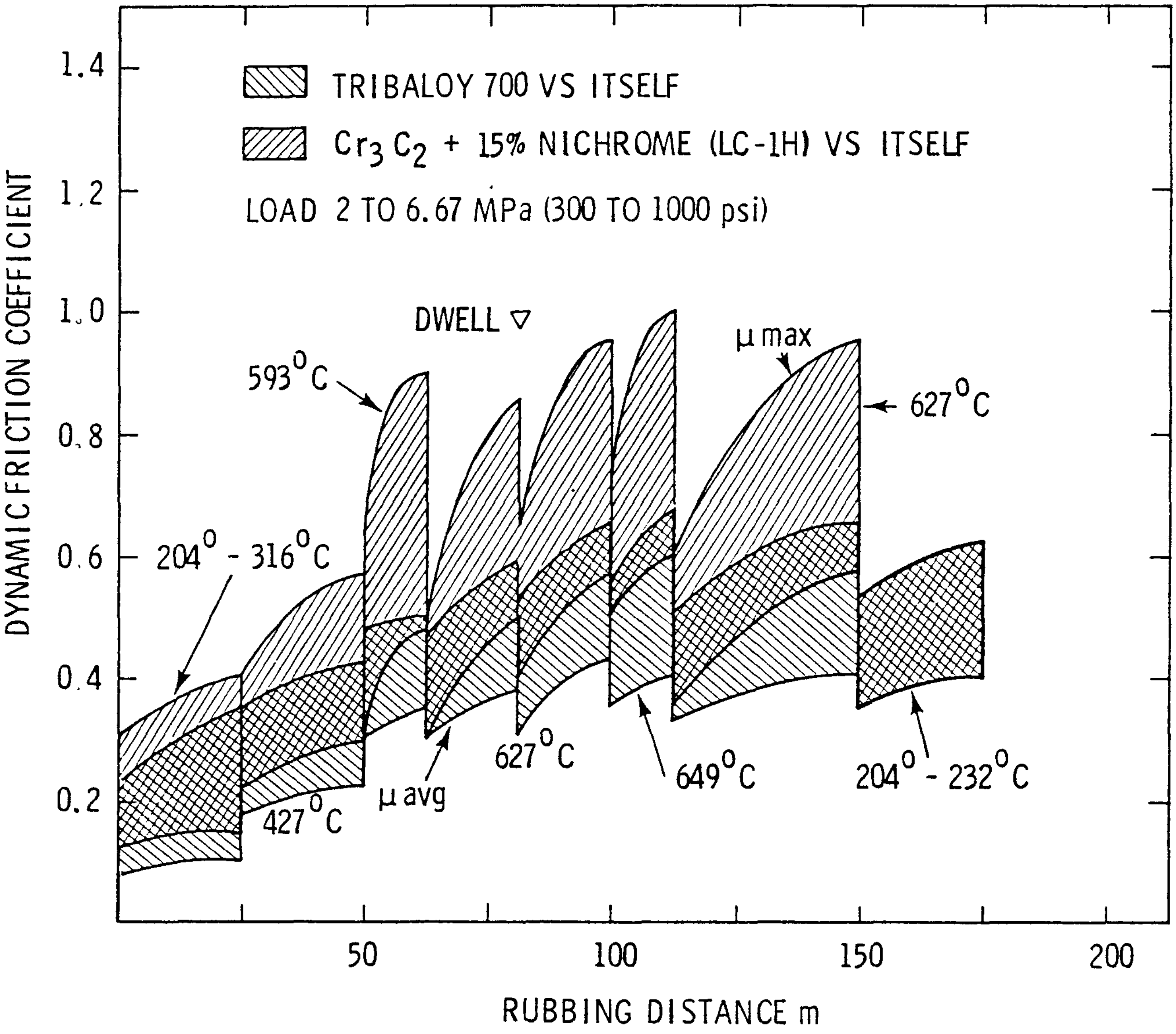

Figure 4 - Friction Comparisons of Tribaloy 700 and Chromium Carbide Detonation Gun Coatings in Sodium.

## NICKEL-ALUMINIDE DIFFUSION COATINGS (ALUMINIZING)

Friction and wear tests were conducted on nickel-aluminide diffusion coated Inconel 718 rubbed against itself in 200°C to 649°C sodium at loads from 0.69 to 33.3 MPa. The results show a reduction in dynamic friction coefficient of 50% or more, when compared to uncoated Inconel 718 tested under the same conditions,(1) as illustrated in Figure 5. In addition to a significant reduction in dynamic friction, the aluminized samples exhibited improved wear and galling resistance. Inconel 718, like Type 316 stainless steel, exhibits adhesive wear behavior and at times severe surface damage and galling in high temperature sodium(6). When nickel-aluminide diffusion coatings are applied however, wear behavior changes to the more desirable mild abrasive or polishing mode. After nearly 210 m of rubbing in 232°C to 649°C sodium at loads of up to 33.3 MPa, no significant wear has been observed. As with the D-gun coatings, surface damage was limited to only polishing and smoothing of high surface asperities with no significant material removal.

Improvements in self-welding tendencies of coated versus uncoated Inconel 718 were as dramatic as those observed for dynamic friction coefficients. Breakaway friction tests conducted after 1-100 minute dwell periods in 593°C sodium indicate a significant reduction in self-welding behavior for coated Inconel 718 when compared to uncoated, as shown in Figure 6.

Similar tests performed on samples with only one half of the wear couple coated (i.e. aluminized Inconel 718 rubbed against bare Inconel 718) also indicate significant reduction in friction coefficients, wear rates, and self-welding tendencies in high temperature sodium. In fact, coating one wear surface appears to be as effective as coating both surfaces.

Nickel aluminide diffusion coatings are non-porous and can be used as corrosion barriers. Sodium compatability tests in 627°C flowing sodium for exposures up to 8000 hours have indicated corrosion rates of about 0.75 μm/yr, as compared to 6.0 μm/yr for bare Inconel 718 under the same conditions. These coatings have exhibited the best corrosion resistance in 627°C sodium of all the coatings discussed in this paper (Figure 3).

Aluminide diffusion coatings appear to be resistant to damage by neutron irradiation(1). A number of specimens of nickel aluminide coatings on Inconel 718 were irradiated in sodium-filled capsules at temperatures from 450° to 600°C

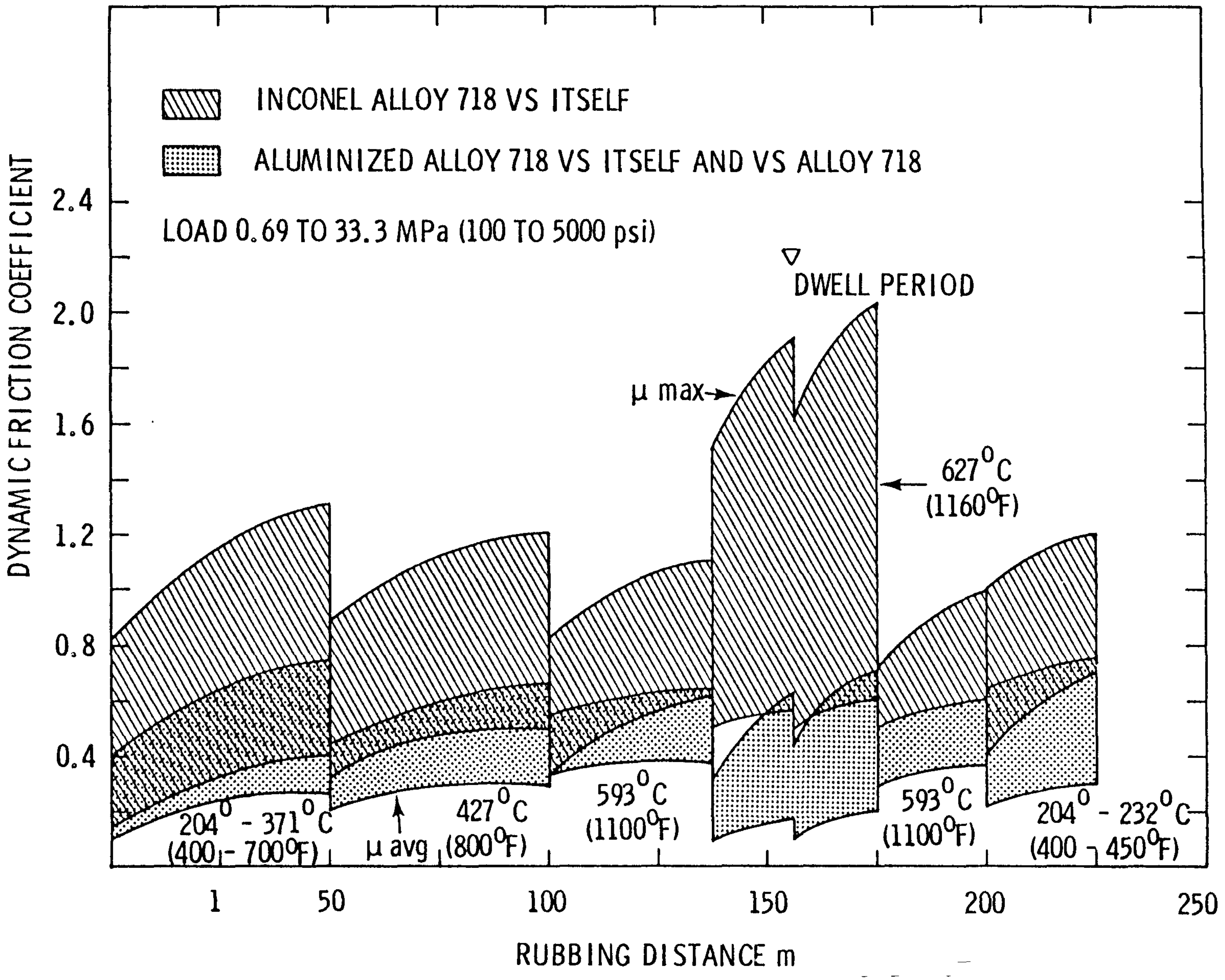

Figure 5 - Effect of Aluminide Diffusion Coating on the Dynamic Friction Coefficient of Inconel Alloy 718 in Sodium.

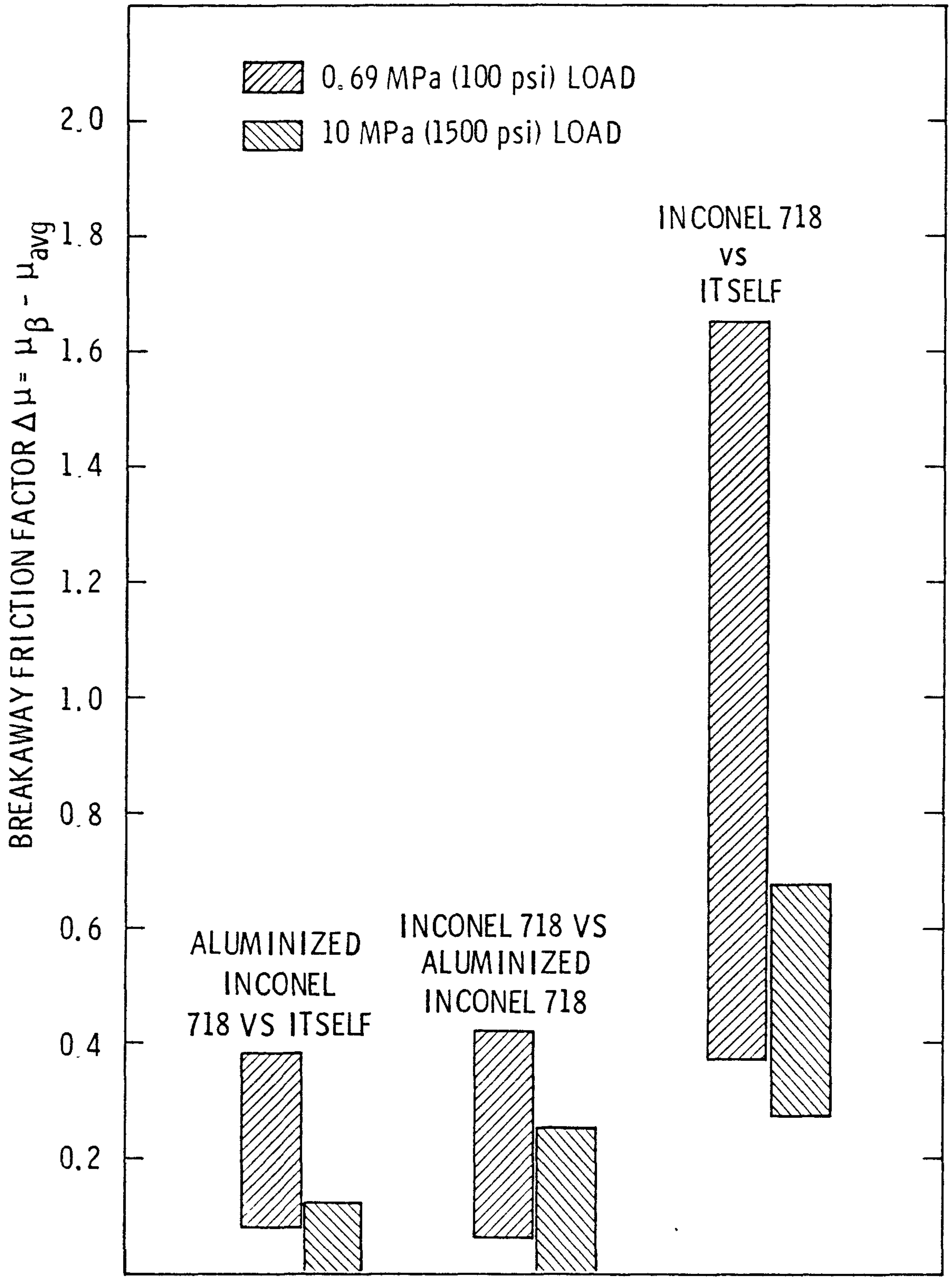

Figure 6 - Effect of Aluminide Diffusion Coating on the Self-Welding of Inconel Alloy 718 in 593° C Sodium.

to total fluences of up to 4 x $10^{22}$ n/$cm^2$. No visible evidence of coating deterioration has been observed. Further irradiation experiments are in progress.

The improvement in friction, wear, and self-welding performance in high temperature sodium resulting from nickel-aluminide diffusion coatings is not restricted to Inconel 718. Coatings have been applied to other structural materials with similar results, as shown in Figure 7. Coatings applied to Type 316 stainless steel have resulted in significant reduction in dynamic friction coefficients, wear rates and self-welding tendency when compared to uncoated material. In addition, the aluminide coating has been the only material tested to date on which Type 316 stainless steel has been rubbed in 627°C sodium without galling.

## SUMMARY

Several chromium carbide detonation gun applied coatings and a nickel-aluminide diffusion coating have been qualified for service in sodium cooled reactors. A Tribaloy 700 and another chromium carbide detonation gun applied coating are nearing qualification status. Successful qualification requires passing friction, wear, self-welding, thermal cycling, corrosion, and irradiation exposure tests conducted under as-prototypic-as-possible service conditions.

Test results indicate that these coatings significantly reduce the dyanmic friction coefficients and self-welding tendencies of Type 316 stainless steel and Inconel 718 in 232° to 649°C sodium. In addition wear behavior is altered from an adhesive mode, accompanied by severe surface damage and galling to an abrasive mode exhibiting little, if any, wear or surface damage.

## ACKNOWLEDGEMENTS

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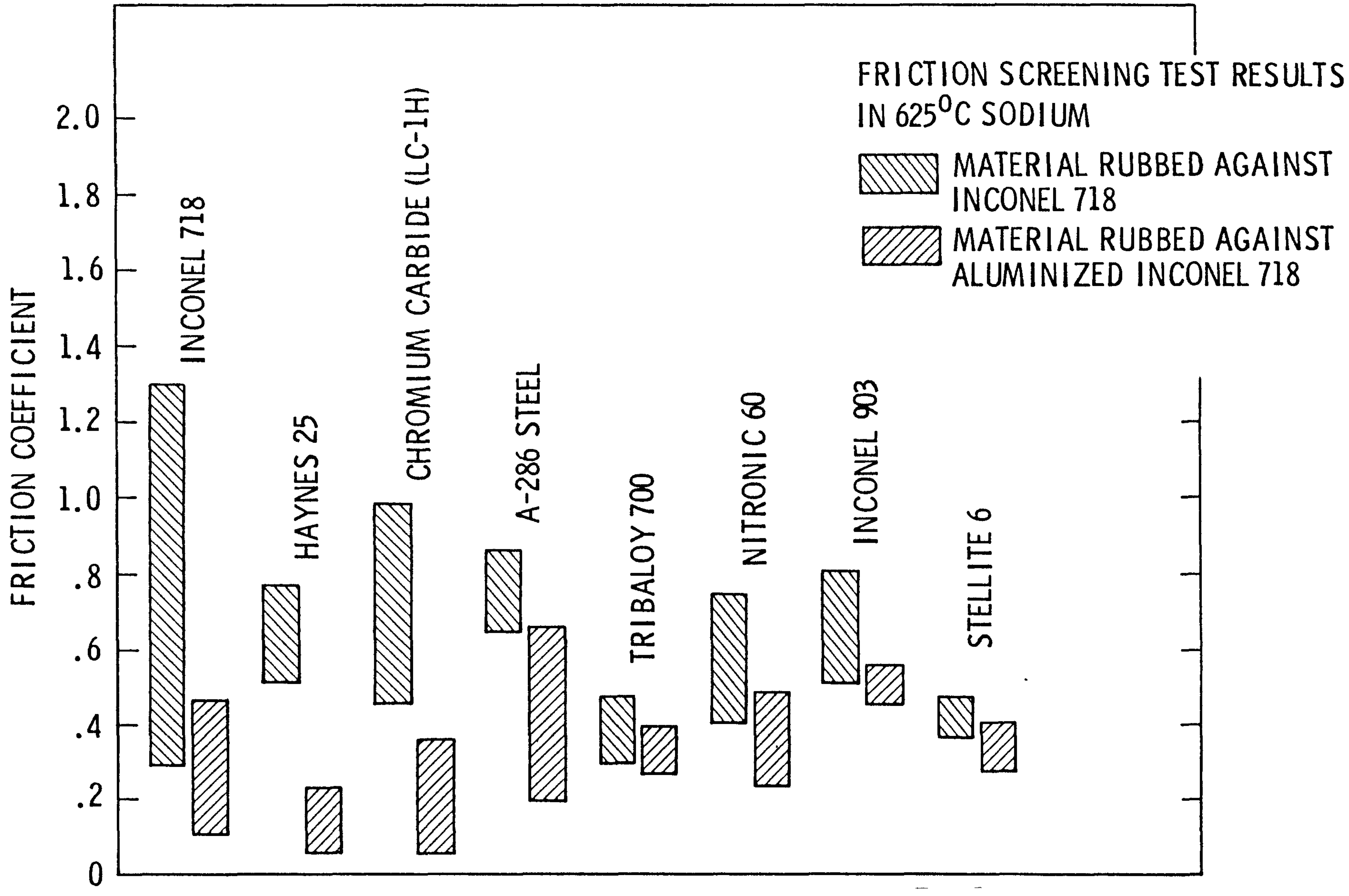

Figure 7 - Effect of Aluminde Diffusion Coating on the Friction of Inconel Alloy 718 Against Other Materials.

Metal Engineering Center) and Rockwell International (RI) performed friction and wear tests and friction screening tests, respectively, in sodium under portions of the program now completed. Particular thanks go to Mr. W. L. Wilson of WARD for his aid and cooperation in the preparation of this paper.

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