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B. R. Seidel, G. L. Batte', N. E. Dodds, G. L. Hofman,
C. E. Lahm, R. G. Pahl, D. L. Porter, H. Tsai and L. C. Walters

Argonne National Laboratory*
Idaho Falls, ID

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B. R. Seidel, G. L. Batté, N. E. Dodds, G. L. Hofman,
C. E. Lahm, R. G. Pahl, D. L. Porter, H. Tsai and L. C. Walters
Argonne National Laboratory
P. O. Box 2528
Idaho Falls, Idaho 83403-2528
(208) 526-7809

ABSTRACT

Significant advances in the understanding of behavior and performance of metallic fuels to high burnup have been achieved over the past four decades. Metallic fuels were the first fuels for liquid-metal-cooled fast reactors (LMR) but in the late 1960's worldwide interest turned toward ceramic fuels before the full potential of metallic fuel could be achieved. Now metallic fuels are recognized as a preferred viable option with regard to safety, integral fuel cycle, waste minimization and deployment economics. This paper reviews the key advances in the last decade and highlights the behavior and performance features which have demonstrated a much greater potential than previously expected.

1. INTRODUCTION

Advances in the development and demonstration of metallic fuels have been both evolutionary and revolutionary in nature over the past decade.

Evolutionary improvements—such as simplifying fabrication; relaxing fuel specifications while maintaining adequate performance; element design improvements; and selection of advanced cladding materials offering minimal irradiation swelling while maintaining ductility and sufficient creep rupture strength—have all contributed to assured reliability to very high burnup under normal and reactor upset conditions while holding down costs.

The revolutionary developments with metallic fuel in the last decade have hinged not on fabrication and performance attributes but upon physical properties which have enabled improvements in reactor safety, an economical fuel cycle and an effective means of consuming actinides rather than discharging them to the waste stream with long-term consequences.

Metallic fuel properties have provided for the significant breakthroughs achieved recently. The high atom density of metallic fuel provides for a good neutron economy and effective breeding. This attribute may not appear significant today when energy resources seem plentiful but it does provide a long-term solution if one is needed and at no additional development cost.[1] The immediate benefit of this property, however, enables minimizing the burnup reactivity swing. This drastically reduces the required control worth such that even with a multiple-rod-runout transient overpower initiator, the passive reactor response limits temperature increase.[2-4]

Another metallic fuel property, high thermal conductivity, enables passive response to the loss-of-flow and loss-of-heat sink upset events. Because of the high thermal conductivity, radial temperature gradients in the fuel are small and heat stored in the fuel is small compared to ceramic fuel. When released, this heat is neither sufficient to elevate the primary coolant more than about 150°C nor challenge temperature limits of structural components.[2-4]

In addition to these properties which influence reactor response, the metallic fuel form enables simple one-step reprocessing by electrorefining.[5] This process removes most of the fission products and carries the transuranic elements to the product. The cathode product can then be easily injection cast into finished fuel slugs, encapsulated in stainless steel tubing and returned to the reactor. Since the process is very direct and metal based, the capital and operating costs for the fuel cycle facility are much less than for an aqueous reprocessing facility.

Another revolutionary breakthrough, however, comes from the propensity of the minor actinides to be inseparable from the uranium and plutonium by electrorefining when reprocessed. This inherent property enables the actinides to be recycled to the reactor where they are fissioned to generate power. With quantitative removal of these long-lived isotopes from the waste stream, the stringent requirements for long-term storage in repositories may be relieved.

The reactor safety features dependent on metallic fuel have already been demonstrated[6-7] at Experimental Breeder Reactor II (EBR-II). Next year, the integral fuel cycle facility adjacent to EBR-II will begin demonstration of the reprocessing and refabrication of metallic fuel. At that time, the reactor system with integral fuel cycle based on metallic fuel can be evaluated as to how well it meets[1,8] all the key acceptance criteria: resource utilization, total cost, inherent safety characteristics, licensability, minimal waste impact, decreased environmental challenges, acceptable deployment scenarios, safeguards, non-proliferation and transportation to mention just a few.

None of these improvements would be possible if it weren't for the good, consistent behavior of metallic fuel. Although most of the characteristics of metallic fuel behavior now under investigation were initially discovered in the late 1960's, recent irradiations and tests have confirmed the consistency of fuel behavior and performance for all metallic alloys, including plutonium-bearing alloys.

2. RECENT DEVELOPMENTS

In 1983, a concept emerged at Argonne National Laboratory, called the Integral Fast Reactor Concept (IFR), the objective of which was to offer a safe and economical solution to the technical and institutional issues that had inhibited nuclear power from meeting a larger share of the world's energy demands.[1,8] Central to the concept was the recognition that the world's reserve of U^{238} must be utilized as an energy source in the centuries to come. Thus, the fuel system must contain plutonium and further the reactor must have good breeding characteristics.

The U-Pu-Zr metallic fuel system, which was under development in the late 1960's, was chosen as the candidate fuel because of superior performance characteristics over other metallic fuel systems. During the period of development in the 1970's at EBR-II, the perceived performance disadvantages of metallic fuel were satisfactorily resolved and as well additional attributes of metallic fuel were discovered. Developments associated with the performance of metallic fuels over the past 30 years have been reviewed in several recent papers.[9-13]

3. IRRADIATION TESTING

Since 1985, nearly a thousand experimental metallic fuel elements covering a range of fuel compositions, design variations and operating conditions have been irradiated in EBR-II and about another thousand in the Fast Flux Test Facility (FFTF). These experimental test elements include operation at two-sigma operating temperatures (660°C), high fuel-smeared density (85-90%), and high fuel-to-plenum volumes (0.7) which severely challenge element reliability; however in every case, the lifetime was greater than expected. Other than some unexpected closure-weld failures which are believed to be prevented now by design changes, breach is only observed at about 16 at.% burnup or higher in aggressively-designed elements; several of these have survived to 18.4 at.% burnup.

The core of EBR-II was completely converted to U-Zr/U-Pu-Zr alloy fuel more than a year ago to statistically demonstrate reliability and performance; several experimental lead elements, including those clad with HT9, have now reached 16 at.% burnup without breach and no performance limitations have been identified. Examination of these elements have characterized the behavior and performance of metallic fuel including fuel swelling, gas release, compatibility with the cladding, breach characteristics and benign operation beyond cladding breach.

Renewed testing began when three complete assemblies of advanced metallic fuel were placed in the core of the EBR-II in February of 1985. The 61-pin assemblies each contained an identical complement of metallic fuel consisting of three compositions in weight percent: U-10Zr, U-8Pu-10Zr, and U-19Pu-10Zr. The pins were clad with the austenitic alloy D9, had a peak linear power rating of 48 kW/m, and achieved peak cladding temperatures of 583°C. The highest burnup achieved on these pins was 18.4 at.% with one end-of-life fuel column failure at 16.4 at.% burnup. These lead assemblies, X419-X421, have demonstrated that metallic fuels have the potential of being competitive with any existing fuel type in terms of steady-state and transient performance.[14-17]

Subsequent to the initiation of the irradiation of the three lead assemblies, a broad-based development program was instituted to fully explore the potential of metallic fuel. To date more than 40 assemblies of experimental fuel have been irradiated with another five being fabricated. Among these were tests characterizing the influence of fabrication variables of minor impurity concentrations and defects in the fuel; cladding defects; design variables of major composition, fuel-smeared density, fuel element diameter, fuel column length and fuel-to-plenum volume ratio; and run beyond cladding breach (RBCB) of both intentionally-defected pre-irradiated elements to accelerate breach and naturally-occuring breaches.[18-19]

Ex-core tests and analyses were initiated to study phase relationships, fuel/cladding compatibility, zone formation, fuel plastic flow properties, thermal conductivity, and thermal expansion. Results from this work as well as the results from the postirradiation examination of the in-reactor experiments are used in fuel performance codes called LIFE-METAL and FPIN for steady-state and transient conditions, respectively.[26-27]

4. IRRADIATION PERFORMANCE RESULTS

A great deal of progress has been made toward the complete understanding of metallic fuel systems. Although most of the phenomena now under investigation were initially discovered in the late 1960's, the recent irradiations have confirmed the consistency of fuel swelling, gas release, compatibility with the cladding and element redistribution within the fuel.[12]

Performance issues important to design of advanced reactor systems include fission gas release, axial growth of the fuel and the breach characteristics demonstrated by metallic fuels.[14-16] The fission gas release behavior is important as it has been found that primary loading of the cladding is produced by fission gas pressure; other loading mechanisms, such as fuel-cladding mechanical interaction, do not appear to be significant until very high burnup is achieved. The molar quantity of gas released is nearly linear with burnup while the fractional release asymptotically approaches 80% theoretical at about 18 at.% burnup, Figure 1.

In general, most of the important phenomena studied show a dependence on plutonium concentration in the U-Pu-Zr alloy. For example, as the fuel is irradiated it swells both axially and radially until contact is made with the cladding. Fuel/cladding contact is complete by 2 at.% burnup and may open again at high exposures with cladding which swells significantly. Radial fuel growth, which is freely promoted in the 73% smear density design, dominates the swelling process until cladding contact at ~2 at.% burnup, while axial growth (which depends on Pu content) is limited to minimal growth after 2 at.% burnup, Figure 2. The extent of axial swelling is a strong function of plutonium concentration with the extent of axial elongation minimal with a plutonium content of 19 wt % at nominal operating conditions.[21] The variance in swelling behavior between U-10Zr and U-19Pu-10Zr vanishes at lower fuel operating temperatures. Microstructural examinations have shown that the swelling mechanism changes as a function of composition and temperature (tearing vs. bubble formation), and the microstructure correlates well with observed degrees of swelling anisotropy.

One of the more interesting phenomena is the radial redistribution of the fuel alloying elements during irradiation.[22] By 2 at.% burnup, an interchange between zirconium and uranium occurs. The plutonium concentration profile remains unchanged. Depending on fuel temperatures and plutonium content, this leaves either a low zirconium (<2 wt.%) shell at mid-radius surrounding a zirconium-rich core or a zirconium-depleted zone at slug center. Pie-shaped cracks in the higher plutonium-content alloys are common at the earliest stages of redistribution but are completely "healed" by fuel growth before 10 at.% burnup. Postirradiation examination is now underway to measure composition profiles at 17-18 at.% burnup. We have found recently that the extent of radial redistribution depends on the alloy composition; as the plutonium concentration of the alloy is increased, the radial redistribution becomes more pronounced. An adequate model for this phenomenon is yet to emerge; the basis must include the results of ex-core diffusion couples and ternary diffusion analysis as well as in-core irradiation observations. To date we have found that the radial redistribution of fuel species does not limit the performance of metallic fuel.

5. BREACH CHARACTERISTICS

Thirteen breaches were obtained from lead assemblies in the range of 13.5 to 18.4 at.% peak burnup. Nine of these 13 breaches were associated with weld failures. The weld failures were all of a benign nature, with no fuel loss nor delayed neutron signals present. These early weld-related failures were clearly unrelated to fuel type.[15]Three other breaches also occurred above the core region and can be considered unrelated to the fuel since the failures were in the plenum region ~100-150 mm above the top of the fuel. Because this is a region of low cladding temperature and strain with no adjacent fuel we can only now speculate that defects induced from remote reconstitution may be involved.[15]

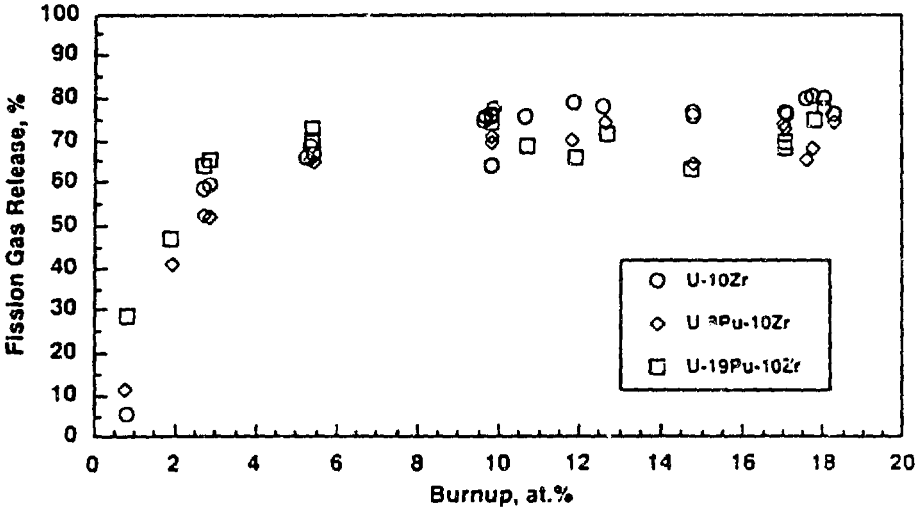

Figure 1. Gas Release from Metallic Fuel[13]

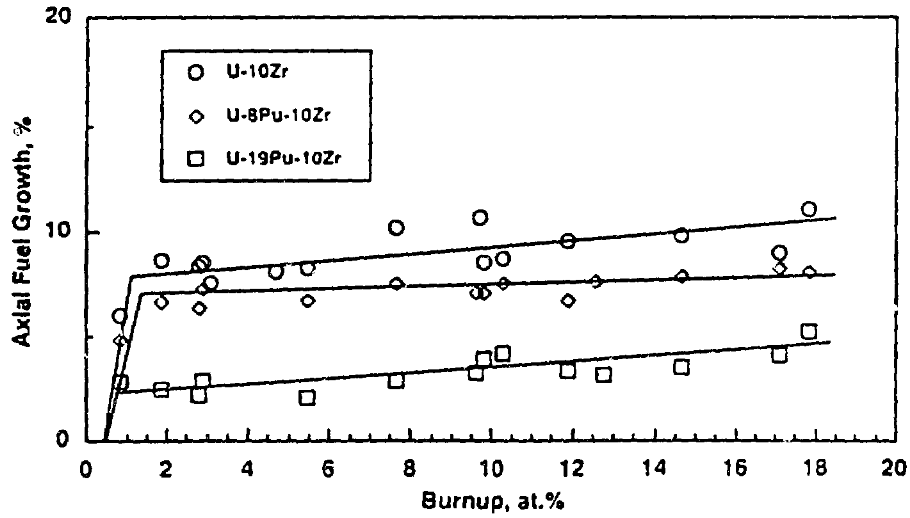

Figure 2. Axial Elongation of Metallic Fuel[13]

The only breach observed that may truly be related to the fuel occurred in element T084 from the X420 test at 16.4 at.% burnup. Exact knowledge of the breach burnup can be made since tag gas signals and delayed neutron signals were coincident. The failure site was in the fuel region of this U-19Pu-10Zr element, ~225 mm from the bottom of the fuel column ($X/L_0 = 0.67$). Expulsion of bond sodium and fission products led to a burst-like DN signal of 20 minute duration approximately four times background. The crack appeared to be very small and tight and was at the azimuthal position of maximum element-element interaction due to the excessive swelling and creep of the cladding at that burnup. Nondestructive examination (neutron radiography and gamma spectrometry) gave no evidence of abnormal fuel restructuring at the breach site but cladding profilometry by laser scanning showed pronounced ovality of the cladding at this elevation. Weight loss analysis showed significant fission gas, bond sodium and liquid fission product venting had occurred. EBR-II operated with this natural fuel column breach for an additional 34 days (0.6 at.% burnup) to the scheduled end of the cycle without consequence.[15]

6. RUN BEYOND CLADDING BREACH PERFORMANCE

The RBCB performance testing was accelerated by pre-thinning the cladding of a preirradiated test element and allowing it to breach under the stress induced by internal fission gas pressure during further irradiation. Several tests were conducted using different ternary fuel alloys of U-xPu-10Zr (x = 0-19 wt %) with Type-316 stainless steel, D9 and HT9 cladding materials. The test pins were previously irradiated to a range of burnups between 3 and 12 at.% prior to thinning the cladding.

Steady-state irradiation and subsequent examination of the RBCB tests have resulted in a large data base that has produced strong support of the fuel's predicted benign behavior under breach conditions.[18-19] Table I provides a description of the RBCB program and its current status to date.

In all of the IFR metal fuel scoping tests, the breach results appeared to have a common characteristic release signature. Typically, upon onset of the breach, an increase in reactor cover gas activity as well as a relatively short-lived delayed neutron (DN) signal was observed. In the case of tagged elements, the tag was readily identifiable. In the case of XY-24 and XY-27, a series of small peaks of activity in the reactor cover gas from fission gas released through the breach was observed. None of the breaches provided any DN activity of a prolonged nature nor a very high DN signal.

Calculated time-to-rupture values using internal gas pressure alone appears to be conservative, potentially due to the bonding of the fuel to the cladding. This metallurgical interaction layer may distribute the stress at the deformation site, thereby delaying the instability that normally occurs in the final stages of creep rupture. Examination of breached elements revealed that no crack widening or fuel loss had occurred during prolonged RBCB operation. Only during the initial breach was there any evidence of significant release of activity from the fuel element. After the release of bond sodium, cesium and other liquid fission products, along with the accumulated fission gas, the reactor cover gas activity ceased, with the exception of periodic small releases of gas.

Based on the results of the scoping tests, a more aggressive series of RBCB experiments were conducted on elements of IFR prototypic design. All three of the prototypic IFR fuel experiments were to be irradiated for approximately two reactor cycles each and consisted of U-10Zr and U-19Pu-10Zr fuel with D9 and HT9 cladding.

The prototypic tests performed thus far have demonstrated the same characteristic pattern as the scoping tests which have also been confirmed by the characteristics of the naturally-occuring breach in X420. In experiments X482 and X482A, short duration DN signals of 600 to 700 cps above background were observed. No further opening of the defect after initial breach occurred and fuel loss was negligible. Although many examinations remain to be performed on the prototypic test elements, the data obtained thus far, taken with the scoping test results, indicate an overall very benign RBCB behavior of IFR metallic fuel.[18-19]

7. LIQUID-PHASE ATTACK AT ELEVATED TEMPERATURE

Another area of intense investigation is the determination of temperature and times for the first appearance of liquid phases in the fuel and cladding interaction zone when exposed to temperatures above nominal steady-state conditions.[13,18-19,23] Laboratory testing of irradiated fuel element sections and whole elements have shown that independent of the fuel alloy composition and cladding material, ferritic or austenitic steel, the temperature for the first appearance of a liquid phase in irradiated samples is above 700°C.[24-25]

Furthermore, thermally-activated cladding penetration rates are low near the temperature for first appearance of a liquid phase. In addition, iterstitial impurities in the fuel and cladding play an important role in the stabilization of a zirconium layer on the periphery of the fuel which tends to retard the interdiffusion of the fuel and cladding at the interface. At high burnup for the higher-swelling D9 cladding which has swelled away from the fuel, the lanthanide fission products are found to segregate near the cladding inner surface and play a major role in compatibility at elevated temperatures. For the D9 cladding, interdiffusion of cladding components, primarily nickel and iron into the fuel and lanthanide fission products out of the fuel into the cladding during steady-state conditions promotes formation of a liquid phase at elevated temperature. After one hour at 800°C, the cladding wall was reduced as much as 26% in U-19Pu-10 Zr fuel clad with D9.

In-reactor demonstration of reliability of metal fuel elements at temperatures above the eutectic formation temperature began with the

Table I. Status of RBCB Testing

	Scoping Tests			IFR Prototypic		
Experiment	XY-21/21A	XY-24	XY-27	X482	X482A	X482B
Composition, wt%	U-5Fs	U-19Pu-10Zr	U-8Pu-10Zr	U-19Pu-10Zr	U-10Zr	U-19Pu-10Zr
Cladding Material	316SS	316SS	316SS	D9	D9	HT9
Initial Burnup, at.%	~9.3	~7.5	~6.0	~11.9	~11.9	~10.6
Element Diameter, mm	4.4	4.4	4.4	5.8	5.8	5.8
Breached Condition, Days	54	233	131	168	~100	—
MWD	3348	18453	15234	9200	6200	—
RUNS	136-139	143-146	144-146	149-150	152-153	154-155
STATUS	NN41(XY21) no breach; RT95(XY21A) breached	J507 breached; J516 no breach	J432 breached; J486 breached	T139 breached	T045 breached	breach upon startup for element T464 from S/A X425A
DN Signal, cps*	~30-40	**	**	~600	~700	—
Weight Loss, g***	~2.0 g	2.7 g	~2.5	4.04 g	not available	—
Irradiation Facility	BFTF	FFTF	BFTF	Open Core	Open Core	Open Core

*counts above background
**unavailable due to malfunction of instrument sensitivity
***expulsion of bond sodium, cesium and fission gas accounts for majority of weight loss; there was negligible fuel loss

EBR-II Mark-II element design consisting of U-Fs fuel and Type 316 stainless steel cladding. Mark-II metallic fuel elements, which only exhibit liquid phase attack at temperatures above 715°C, were operated at temperatures up to 800°C in EBR-II in a 61-element subassembly, XY-22, in order to characterize high temperature breach.[28] Fuel elements with a large range of burnups were included in the test in order to compare the damage caused by the over-temperature operation at various stages in fuel element life. The subassembly operated at elevated temperature for ~42 minutes when failure of a high burnup element (7.69 at.%) occurred and the test was terminated. The failure was identified as being due to stress rupture at the fuel restrainer dimple (a small indentation in the cladding designed to limit axial fuel growth or motion) and both mode and time-to-failure agreed very well with pre-test predictions. The unbreached elements, having sustained some limited damage to the cladding due to liquid phase formation, were removed and irradiated in another subassembly (X427) at normal operating temperatures until first breach. Experiment X427 contained all the remaining elements from the in-reactor eutectic penetration tests and enough fresh elements to fill a 91-element bundle.

The XY-22 test indicated that low-burnup elements had more cladding attack than the high-burnup elements. The high-burnup elements failed due to stress rupture in the restrainer dimple common to normal end-of-life failure. It was expected that the low-burnup elements could breach early due to thinning of the cladding in the fuel region prior to the normal dimple failure, so all elements were irradiated at the same time to determine which would fail first. Breach was detected after an increment of ~2 at.% burnup beyond the initial high-temperature test. Two of the high burnup elements, at 10.0 at.% and 10.2 at.% burnup, both higher than the administrative burnup limit of 8 at.% for the Mark-II design, exhibited a weight loss normally associated with breach. This indicated that the normal end-of-life failure mode was to be expected for the high-burnup elements, even after an extended over-temperature event. It was also necessary to determine if the mode of failure changed as a function of burnup. All the medium to high burnup elements were then removed from the subassembly and the low burnup elements were allowed to continue irradiation as X427A until the nominal end-of-life was exceeded. The final burnup for these elements was >11 at.% when the test was terminated without breach.

The X427-X427A tests demonstrated that damage to the cladding due to short term over-temperature events would not significantly reduce the lifetime of these metallic fueled elements.[13-23] The first failure of a high-burnup element occurred at 10.0 at.% burnup, which is within the 2-σ band for the expected lifetime. *Post-irradiation examination* of the elements showed evidence of previous high temperature attack during the XY-22 irradiation, but no significant cladding degradation. These elements had fuel/clad interaction layers of limited thickness and no significant thinning of the cladding was evident.

Although no fuel failures occurred in the elements exposed to high temperature at low burnup and subsequently irradiated to high burnup, the fuel restrainer dimple in these elements was found to be the weakest point. The dimple acts as a stress riser for both steady-state and over-temperature events. Fuel elements currently used in EBR-II, and proposed for IFR-type reactors, have no fuel restrainer dimples and therefore may be ultimately limited by the cladding wastage incurred during over-temperature events. Design of advanced reactor fuel elements has indicated that predicted stress rupture failure during anticipated over-temperature events at end-of-life may be used to set exposure limits and that limited liquid phase attack may be important only at beginning-of-life when significant thinning of the cladding could occur before failure.

8. SUMMARY

All test results have demonstrated the robust behavior of metallic fuel and it's potential to achieve high burnup even with aggressively-designed elements. The key characteristics of behavior are now more clearly understood and no limitations to IFR applications have been identified.

The direction of the fuel performance program is now shifting toward the generation of a statistically significant data base to determine whole-core performance of the advanced metallic fuel. The core of EBR-II has been converted to U-10Zr with limited U-xPu-10Zr, where x varies between 8 and 28 wt % plutonium. This conversion will not only offer the opportunity to gain data on large numbers of assemblies, but this fuel will also be the first material introduced into the new IFR reprocessing facility currently under construction at Argonne National Laboratory near Idaho Falls.

Much work remains before metallic fuel can be considered fully licensable. Results to date, however, show that metallic fuel is readily and economically fabricated, is capable of achieving high exposures and long reactor residence times, and possesses unique and promising safety features.

9. ACKNOWLEDGEMENTS

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