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Densification Testing
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Densification Testing

Licensing Topical Report

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DISCLAIMER OF RESPONSIBILITY

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REFERENCES

1) R.O. Meyer, The Analysis of Fuel Densification, NUREG-0085, July 1976

2) USNRC Regulatory Guide 1.126, An Acceptable Model and Related Statistical Methods for the Analysis of Fuel Densification, Rev. 1, March 1978

3) M.O. Marlowe, In-Reactor Densification Behavior of UO_2, NEDO-12440, July 1973

Densification Testing

1. INTRODUCTION

Since the discovery of in-reactor densification of oxide nuclear fuels in 1972, the impact of that densification on safety related issues has been routinely considered in fuel design and fabrication. Those considerations include the effects of densification on linear heat generation rate due to shortening of the fuel pellet column, stored energy in the fuel due to an increase in the fuel-cladding gap, power peaking due to potential formation of axial gaps in the fuel pellet column, and the possibility of power spiking due to creep collapse of the cladding over those regions of the fuel where axial gaps in the column have occurred and the resulting increase in local moderation.

The extent and kinetics of the in-reactor densification were found to be related to the densification that occurred on thermal treatment of the fuel pellets at 1700°C for 24 hours.[1,2] Routine testing of production fuel pellets has been continued by GE/GNF at a high frequency over the ensuing 30+ years since that time.

Since the recognition of in-reactor densification, the design and fabrication of the fuel have advanced significantly to achieve improved fuel cycle economics and performance, and to provide reduced product variability during manufacture. It will be demonstrated in this report that these factors have served to reduce the mean and minimize the variance of the densification observed by thermal simulation at 1700°C for 24 hours.

Therefore, with these advances in fuel design and fabrication, the current high frequency of densification testing of production fuel is no longer required.

2. FUEL PELLET DESIGN HISTORY

Since the 1970's, the need for improved fuel cycle economics without reduction in reliability or safety has driven numerous changes in BWR fuel design. Some of those changes include modified fuel bundle lattice configurations, i.e., 7x7 → 8x8 → 9x9, with successively decreased fuel pellet diameter and stored energy per pellet during operation, and the addition of pre-pressurization of the fuel-clad gap with helium to improve fuel-clad gap thermal conductance and dilute the detrimental effects of fission gas release thereon.

The fuel pellet design has also been modified to achieve the same objectives. In particular, the pellet density has migrated progressively higher to affect greater fuel loadings and capitalize on lower than previously anticipated needs to accommodate fuel swelling and fission gas release.

The history of the fuel pellet density requirements for GE/GNF fuel pellets is shown in Figure 1. Note that the nominal density has increased by [[ ]], from [[ ]] of theoretical density, since the discovery and control of fuel densification. The lower extreme of acceptable densities has increased even more, from [[ ]] for individual pellets to [[ ]] for the lower 95/95-tolerance limit on pellets in a fuel reload project. While we now realize that post-fabrication densification is not solely determined by the as-fabricated, initial density, it is interesting to note that the earliest regulatory requirements for assessment of fuel densification[1] assumed that terminal density was achieved at [[ ]] of theoretical (geometric – which is essentially equivalent to [[ ]] of true theoretical density). By that measure, modern GE/GNF fuel would nominally be immune to in-reactor densification.

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]]Figure 1. History of GE/GNF Fuel Density Requirements

It is generally true, however, that for a given powder fabrication and pelletizing process the propensity for densification is inversely related to the density of the as-sintered pellets. Therefore, the very substantial increase in the lower density limits [[ ]] has acted to strongly alleviate, if not eliminate, concerns about fuel pellets with large densification potentials.

The GE/GNF fuel densification requirement history is shown in Figure 2. By contrast with the progressively increasing density requirements, the densification resistance requirements have remained essentially constant.

Because of the general relationship between density and densification propensity, the increase in pellet density requirements with no increase in densification resistance requirements have combined to substantially enhance the ability of GE/GNF to meet the densification requirements.

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]]Figure 2. History of GE/GNF Densification Requirements

3. FUEL PELLET MANUFACTURING HISTORY and PRACTICE

Several fundamental changes have taken place in UO_2 powder and fuel pellet manufacturing as conducted by GE/GNF since the early 1970's that have led to improved consistency and densification resistance.

4. ENRICHED URANIUM PELLETS

4.1 UO_2 Powder Production

From the 1970's through the mid-1990's UO_2 powder for fuel pellet production was simultaneously produced by GE/GNF using several chemical-processing routes. Those routes included the ADU (ammonium diuranate) process, GECO – a GE proprietary dry conversion process, and a wet chemical recovery process. In addition, there were multiple calcination-options for each of these basic chemical processes as well as options for the addition of volatile pore formers and for cross-blending powders of these various types. This plethora of processes and their attendant differences in densification behavior accentuated the need for densification testing of materials from each process option. In addition, it diluted the statistical strength of the sampling of each powder process type, and contributed in a negative way to the statistically derived densification upper-limit assessments.

The statistical variability was further exacerbated by the use of relatively small UO_2 powder lots, e.g., 350-500 kg.

In 1998, however, there was a transition by GE/GNF to a single UO_2 powder production process, i.e., the DCP (dry conversion process). Attendant with that process change was a change to large, 1500-2200 kg, powder lots. Since that time, that single UO_2 powder type has been used both for UO_2 pellets and $(U,Gd)O_2$ pellets. This singularity of powder types, and the use of large, uniform powder lots has led to a highly uniform sintered density and densification test results across all of the production material. It has also significantly improved the statistical analysis of each powder lot and type.

4.2 Pellet Sintering Conditions

Prior to the discovery of in-reactor fuel densification the control of final pellet sintered density was often achieved by control of the sintering cycle. The final sintered densities were known to be highly dependent on the sintering temperature and time. The maximum sintering temperature used in production and the time at that temperature were adjusted based on results from sample pellets to produce final densities within acceptance limits. Those sintering conditions varied widely. If the powder was highly active, to achieve the final nominal density of [[ ]] of theoretical often required sintering temperatures as low as [[ ]]. Occasionally, volatile pore formers were used to simplify achieving the correct final density with active powders; at that time the use of pore formers was more of an exception rather than routine practice.

Following the discovery of in-reactor densification and implementation of routine densification testing, it was found necessary to increase the sintering temperature/time to adequately assure dimensional stability of the sintered pellets. During the later 1970's and early 1980's, the GE/GNF sintering conditions migrated to the conditions that are currently used, i.e., [[ ]]. Note that the 'heat-work' input to the production material, as measured by the product of the volume diffusion coefficient, which defines the rate of density increase during sintering, and the sintering time[3], or D x t, increased by a factor of as much as 13.

Beginning by about 1981, the density/densification control philosophy at GE/GNF had changed to the utilization of constant [[ ]], high-temperature [[ ]] sintering for all powder types. Final density control was achieved by the use of volatile pore formers, for example ammonium oxalate (AO). The concentration of pore former needed in each powder lot was determined based on sample pellets from that lot and sinterability tests.

With the migration of the nominal density upwards in the early 1980's and again in 2000 [[ ]], pore formers are used to a lesser extent. However, with the transition to the highly active DCP UO_2 powder in 1998, a volatile pore former is still widely used to achieve the design density requirements in conjunction with incorporation of dry recycle U_3O_8, which also has a small pore-forming effect.

This overall philosophy of using highly active DCP powder, large uniform powder lots, constant high temperature sintering conditions that serve to stabilize the microstructure, and final density control by the use of pore formers has produced fuel pellets with consistently low densification.

4.3 Fuel Pellet Density – Quality Control

Because of the importance of the final pellet density in achieving fuel design objectives including economics, safety, and reliability, and the sensitivity of the final pellet density as an indicator of the extent of control of the powder and pellet production processes, high levels of sampling of fuel pellet density are maintained. In addition, because of the importance of fuel density to economics and safety, redundant 100% screening is provided at the point of final pellet loading into fuel rods.

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These quality control practices provide great assurance that the density of the pellets is within specification requirements. The high frequency of testing and multiple over-checking provide a high level of assurance that low-density pellets, which might be susceptible to greater-than-anticipated densification and thus in turn might affect fuel rod performance, will not be inadvertently included in assembled fuel.

5. NATURAL U – UO_2 PELLETS

Since 1994, UO_2 pellets with natural ^{235}U content have been obtained by GE/GNF from Canadian General Electric (CGE) in Toronto, Canada. CGE fabricates fuel containing only natural ^{235}U for the heavy-water-moderated CANDU reactors in Canada and elsewhere. Because of the unique nuclear characteristics of the CANDU design and the limited burnup achievable, there has always been a premium on high-pellet density. High-density pellets, e.g., with nominal density in excess of [[97.0%{3}]] of theoretical, have historically been the standard for the CANDU reactor applications. Therefore the density and densification resistance requirements for GE/GNF BWR natural ^{235}U fuel were well within the established process capability and routine practice for CGE.

6. FUEL PELLET DENSIFICATION HISTORY and PRACTICE

Historical GE/GNF fuel pellet densification performance from 1994 through 2003 relative to specified requirements is shown in Figures 3 through 8.

In Figure 3, the upper 95% confidence levels on the mean pellet densification are shown for the major enriched UO_2 powder types produced by GE/GNF. These yearly-average values were obtained from rolling statistics on project quantities of material produced by GE/GNF for each pellet type over the course of each year. Also shown for each year in Figure 3 is the number of standard deviations between the upper 95% confidence level on the mean densification and the specified limit.

In Figure 4, similar information is provided as in Figure 3, but for $(U,Gd)O_2$ pellet types produced by GE/GNF.

In Figure 5, similar information is provided as in Figure 3, but for pellets with natural ^{235}U content produced by CGE.

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]]Figure 3. Yearly Average Upper 95% Confidence Level on Mean Densification and the Number of Standard Deviations to Specified Limit for Enriched UO_2 Pellet Types

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]]Figure 4. Yearly Average Upper 95% Confidence Level on Mean Densification and the Number of Standard Deviations to Specified Limit for Enriched $(U,Gd)O_2$ Pellet Types

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]]Figure 5. Yearly Average Upper 95% Confidence Level on Mean Densification and the Number of Standard Deviations to Specified Limit for Natural ^{235}U (CGE) Pellets

In Figure 6, the upper 95/95 tolerance limits on the individual pellet densification are shown for the major enriched UO_2 powder types produced by GE/GNF. These yearly average values were obtained from rolling statistics on project quantities of material produced by GE/GNF from each powder type over the course of each year. Also shown in Figure 6 for each year is the number of standard deviations between the upper tolerance limits on individual pellet densification and the specified limit.

Figures 7 and 8 provide similar information for $(U,Gd)O_2$ pellets and for pellets with natural ^{235}U content produced by CGE, respectively.

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]]Figure 6. Yearly Average Upper 95/95 Tolerance Limits on Individual Pellet Densification and the Number of Standard Deviations to Specified Limit for Enriched UO_2 Pellet Types

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]]Figure 7. Yearly Average Upper 95/95 Tolerance Limits on Individual Pellet Densification and the Number of Standard Deviations to Specified Limit for Enriched $(U,Gd)O_2$ Pellet Types

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]]Figure 8. Yearly Average Upper 95/95 Tolerance Limits on Individual Pellet Densification and the Number of Standard Deviations to Specified Limit for Natural ^{235}U (CGE) Pellets

The data in Figures 3-8 show that during the period from 1994 through 2003 there was always a very large margin for GE/GNF fuel pellet densification to both the mean and individual pellet densification requirements. With the introduction of DCP UO_2 powder for both enriched-UO_2 and $(U,Gd)O_2$ pellet production, that margin made a clear and significant step-wise improvement to larger values. Subsequently, the densification behavior has remained very consistent.

For enriched-UO_2 pellets, both yearly average densification parameters (mean/95 and 95/95) have maintained at least 10 standard deviations from the specified requirements since the introduction of DCP powder. For $(U,Gd)O_2$ pellets, the yearly average of the 95% confidence level on the mean densification has maintained at least 11 standard deviations from the requirement and the yearly average 95/95-tolerance limit on individual pellets has maintained at least 6 standard deviations from the requirement. For natural ^{235}U pellets, the yearly average densification parameters have maintained at least 6 standard deviations from the requirement for the period from 1998 to 2003, and the 95/95 tolerance limit on individual pellets has maintained at least 6 standard deviations from the requirement since the inception of use of CGE pellets.

7. FUTURE FUEL DENSIFICATION CONTROLS

7.1 Routine Densification Testing

By all accepted standards the results described and shown above are indicative of processes that have very good process capability, with virtually no chance of producing out-of-specification material. For example, for a process operating at 6 standard deviations from the requirement in the short term, which is considered comparable to a process capability of 4.5 standard deviations in the long term, the probability of producing an out-of-specification product is less than 4 in a million. The numbers reported in the graphs above are based on long-term data and represent long-term capabilities, thus, the chance of producing pellets with out-of-specification densification resistance from these processes is even less. This consistent process capability for densification behavior of GE/GNF fuel pellets has been maintained since the implementation of the dry conversion process; results have been shown in this report from 1998 to 2003.

If there were to be some major error or excursion in fuel pellet manufacturing (for example a change in sintering conditions, excessive or insufficient pore-former addition, changes in the chemical powder production processes that would lead to a change in powder sintering activity) routine density sampling and the extensive redundant checks on final pellet density would lead to detection of the excursion. Since fuel densification is so closely related to the density of the pellets, these density over-checks also provide protection against any major excursionin densification behavior.

Given the reliable, stable process capability relative to densification resistance, and the routine and redundant 100% checks on fuel density, continued routine densification testing of the GE/GNF fuel pellets is judged to be no longer required to assure acceptable in-reactor densification. Based upon this conclusion, GE/GNF requests the approval of this LTR to eliminate the routine evaluation of the fuel-pellet densification.

7.2 Future Material and Process Changes

The elimination of routine densification testing will require modification of the engineering specifications that define the densification requirements. In order to assure that there is no significant degradation in the densification resistance of the GE/GNF product as a result of any future change in materials or processes, a Qualification Requirement will be added to the specifications for any change in materials or processes that is anticipated to affect densification resistance. Some examples include a change in the UO_2 powder type, a change in the volatile pore former material, or changes in the sintering temperature profile.

It is further planned that the required Qualification testing of proposed materials or process changes will be conducted using a statistical comparison of the extent of densification of pellets from the existing and proposed materials and processes during re-sintering in the production sintering furnaces using the standard (at the time of the change) production sintering cycle. If that statistical comparison shows a statistically significant change in expected densification with the proposed material or process, and Qualification of the new process is still desired, the current standard 24 hour – 1700°C test or an Engineering approved equivalent, will be used to accurately assess the in-reactor densification for consideration in all potentially affected design and licensing analyses.