



Oxidation kinetic changes of UO_2 by additive addition and irradiation

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Received 4 January 1999; accepted 21 June 1999

Abstract

The kinetic changes of air-oxidation of UO_2 by additive addition and irradiation were investigated. Several kinds of specimens, such as unirradiated- UO_2 , simulated- UO_2 for spent PWR fuel (SIMFUEL), unirradiated-Gd-doped UO_2 , irradiated- UO_2 and -Gd-doped UO_2 , were used for these experiments. The oxidation results represented that the kinetic patterns among those samples are remarkably different. It was also revealed that the oxidation kinetics of irradiated- UO_2 seems to be more similar to that of unirradiated-Gd-doped UO_2 than that of SIMFUEL. © 2000 Elsevier Science B.V. All rights reserved.

PACS: 28.41.Kw

1. Introduction

The long-term storage of spent nuclear fuel is becoming a key factor for continuing nuclear power production in most nuclear power generating countries. The storage-term has been considered a longer period than expected before. The reasons are the lack of progress on the repository and the delay of policy decisions of the back end fuel cycle, etc. Dry storage has been regarded as a safe and economical method for long-term storage. However, higher storage temperatures, compared to wet storage, is a weak point. Especially, it may give rise to the severe degradation of defective fuel during the storage period. One of these degradation mechanisms is the oxidation of UO_2 inside the fuel rod by infiltrated air. It has been known that the sequential oxidation of UO_2 in air forms oxygen-rich uraninite derivatives to compositions near U_3O_7 and in turn U_3O_8 [1,2]. After complete oxidation, the pellet form of UO_2 changes to a powder type of U_3O_8 . At this time, about 36% net volume expansion occurs because of the structural change of UO_2 to U_3O_8 , and the expanded volume can

deteriorate the defective fuel rod further. If this occurs during dry storage of fuel, it complicates subsequent handling and storage of the fuel. Therefore, it is important to understand the UO_2 fuel oxidation behavior to define allowable conditions for a safe handling and storage.

The oxidation study of UO_2 has widely been investigated for about 40 yr [3–9]. McEachern and Taylor [10] reviewed extensively the oxidation behavior of UO_2 at temperatures below 400°C and suggested the need for further studies. Many important aspects of the oxidation process are not yet fully understood, and especially it has been reported that there are significant differences in the oxidation kinetic behavior between unirradiated- and irradiated- UO_2 . Therefore, in this study we focus on investigating the kinetic changes of UO_2 oxidation due to some additives, such as Gd and fission product elements, and irradiation. The kinetic changes of these fuels are also compared with each other.

2. Experimental

2.1. Specimens

Four different types of UO_2 were used for these studies.

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(1) *SIMFUEL (SIMulated nuclear FUEL)*. SIMFUEL samples were simulated to 0, 15, 33 and 50 GWd/tU burnup levels by adding different quantities of fission product elements and were fabricated into a pellet type through a 4 hour sintering process in hydrogen at 1700°C. The compositions of the SIMFUELS, which are based on the results calculated by ORIGEN-2.1 code [11], are given in Table 1. The pellet was cut into 1 mm thick disks and polished by 800 grit abrasive paper in order to have a same surface condition. The disks were rinsed, washed with water and dried under room temperature air.

(2) *Unirradiated-Gd-doped UO₂*. Five different types of Gd-doped pellets with 0, 2, 5, 10, and 15 wt% gadolinia contents were fabricated and prepared by the same method used for SIMFUELS. Before the oxidation test, each sample was examined by XRD as indicated in Fig. 3(a). The XRD peak on 15 wt% Gd-doped pellet showed a small splitting caused by a little deficiency in an entire solid solution formation of Gd with the matrix fuel.

(3) *Irradiated-UO₂*. Irradiated-UO₂ samples were taken from spent fuel rods, which were discharged from commercial PWRs and had burnups between 16 and 38 GWd/tU. The burnup measurement was performed by a longitudinal gamma-scanning equipment which has 10 μm scanning precision. Several fragment samples (about 1 g) from each cutting position of fuel rods (section-average burnup position) were selected.

(4) *Irradiated-Gd-doped UO₂*. Irradiated-Gd-doped UO₂ samples were also taken from a spent fuel rod. They had burnups with 10 and 28 GWd/tU. The initial content of gadolinia was 6 wt%. By calculation of the ORIGEN 2.1 code, the residual content of Gd isotopes is 98% of the initial Gd isotopes after irradiation of 28 GWd/tU burnup. This implies that the initial content of Gd in our samples remains almost unchanged.

Table 1
Composition of SIMFUEL (unit: g)

Material	Specimen		
	SFA (15 GWd/tU)	SFB (33 GWd/tU)	SFC (50 GWd/tU)
U	100	100	100
Nd ₂ O ₃	0.2890	0.6357	0.9632
ZrO ₂	0.2174	0.4782	0.7245
MoO ₃	0.2196	0.4831	0.7320
CeO ₂	0.4426	0.9738	1.4754
RuO ₂	0.1891	0.4160	0.6303
BaCO ₃	0.0908	0.1997	0.3026
La ₂ O ₃	0.0650	0.1431	0.2168
PdO	0.0715	0.1574	0.2385
SrO	0.0564	0.1242	0.1882
Y ₂ O ₃	0.0271	0.0597	0.0904
Rh ₂ O ₃	0.0235	0.0518	0.0785

2.2. Experimental methods

The oxidation tests for unirradiated-UO₂ samples with and without additives (SIMFUELS and unirradiated-Gd-doped UO₂) were performed at various temperatures under unlimited air environment by using a cylindrical type furnace. The weight gains were measured by a Cahn-30 micro-balance continuously, and the results were recorded using an automatic data acquisition system. The precision of the micro-balance was 0.1 μg. The unirradiated-Gd-doped UO₂ was also examined by XRD to know the phase changes before and after oxidation.

Irradiated-UO₂ and -Gd-doped UO₂ were oxidized in a hot cell at KAERI's PIEF (Korea Atomic Energy Research Institute's Post Irradiation Examination Facility) at various temperatures and the weight gains were measured by Mettler AE240 micro-balance intermittently. The balance has the precision of 10 μg.

Before oxidation tests, a pretest was performed to know the oxidation kinetic effect by the sample shape difference between irradiated samples (fragment type) and unirradiated ones (disk type). The fragment and disk type samples of unirradiated UO₂ were prepared and oxidized in hot cell and cold laboratory equipment. However, no distinct kinetic difference was found between these two types of fuels.

3. Results

3.1. SIMFUELS

Oxidation tests were carried out at 300–375°C. Fig. 1 shows the test results. Normal UO₂ having no simulated fission products (designated as SF0 in the figures) and all the other SIMFUELS (designated as SFA, SFB, and SFC in the figures) indicate sigmoidal type kinetics at all test temperatures, which are known to appear by a nucleation-and-growth mechanism [3]. The numerical values in parenthesis in the figures indicate the simulated burnup in terms of GWd/tU. The figures also show that the saturation times of SIMFUELS at all test temperatures increase as the simulated burnups increase. Though we could not observe directly the powdering by formation of U₃O₈ during these experiments, from the results it may be suggested that the time of U₃O₈ powder formation (*t_p*) on UO₂ should depend upon the burnup levels.

3.2. Unirradiated-Gd-doped UO₂ fuels

The oxidation test results of 0%, 2%, 5%, 10% and 15% Gd-doped fuels at 350°C are given in Fig. 2. The Gd-undoped UO₂ sample (0%) shows a typical oxidation behavior characterized by a sigmoidal reaction

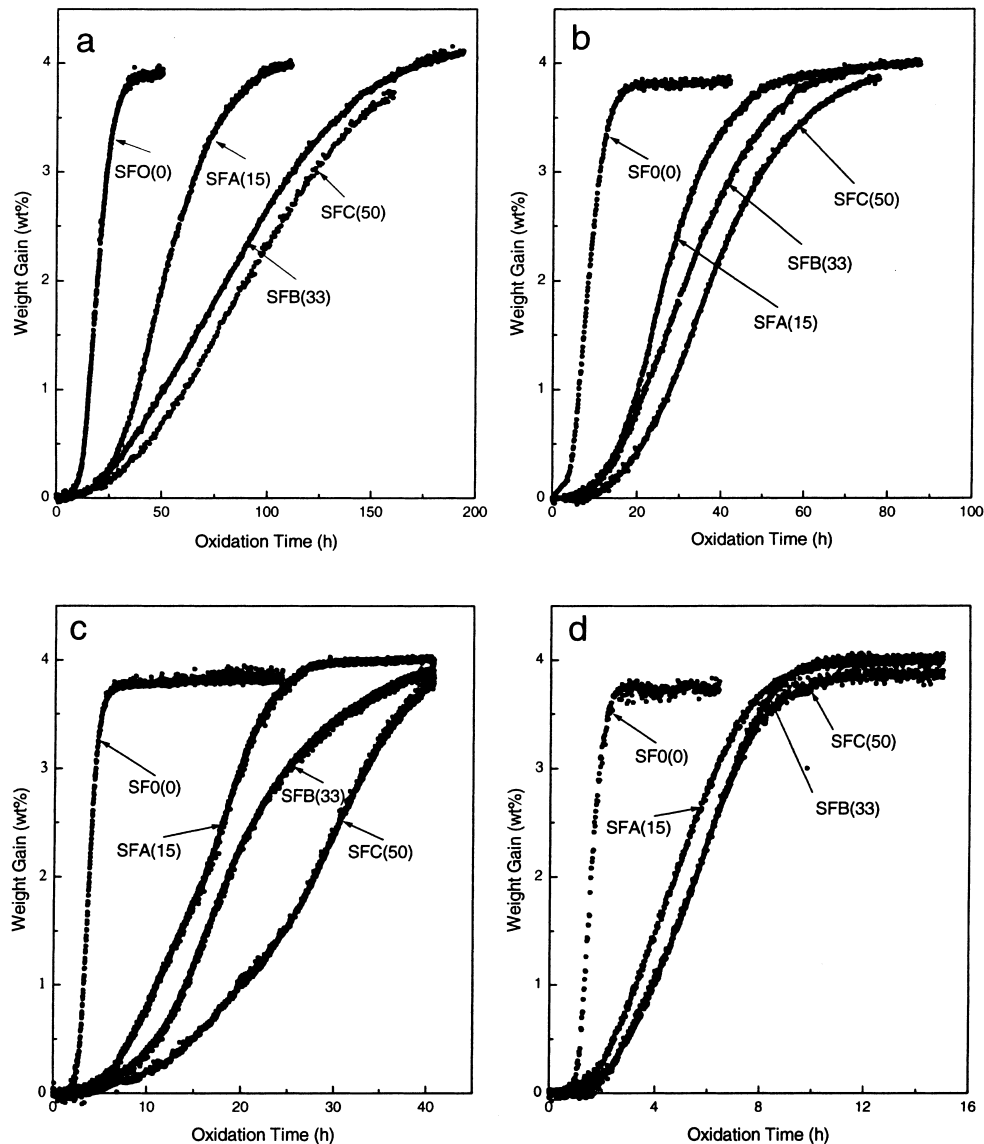


Fig. 1. Oxidation behavior of SIMFUEL. (a) 300°C, (b) 325°C, (c) 350°C and (d) 375°C.

curve and is completely oxidized at about 4% weight gain. However, all the other Gd-doped UO_2 fuels indicate fast oxidation rates at the initial stages, and show slow increases and intermediate-levels of weight gain while continuing oxidation. The intermediate-levels of weight gain decrease with increasing doping quantities. The differences of intermediate-levels of weight gain also increase with the content of gadolinia. After 12-h oxidation, the 2% Gd-doped specimen showed a complete powdering while the 5% specimen showed a localized powdering. However, no powdering was found in the 10% and 15% Gd-doped fuels.

Fig. 3 shows the XRD analysis results of Gd-doped UO_2 fuels with 0%, 5%, 10% and 15% gadolinia contents before Fig. 3(a) and after Fig. 3(b) 15-h oxidation. There are no other peaks except UO_2 on the samples before oxidation, as shown in Fig. 3(a). However, in Fig. 3(b), we can see U_3O_8 and $\text{U}_4\text{O}_9/\text{U}_3\text{O}_7$ peaks in the 5% and 10% Gd-doped UO_2 fuels, but only dominant $\text{U}_4\text{O}_9/\text{U}_3\text{O}_7$ peaks in the 15% Gd-doped UO_2 fuel. These XRD results mean that the dopant levels can affect the nucleation and growth of U_3O_8 on the surface of UO_2 and even inhibit the formation of U_3O_8 at a certain quantity.

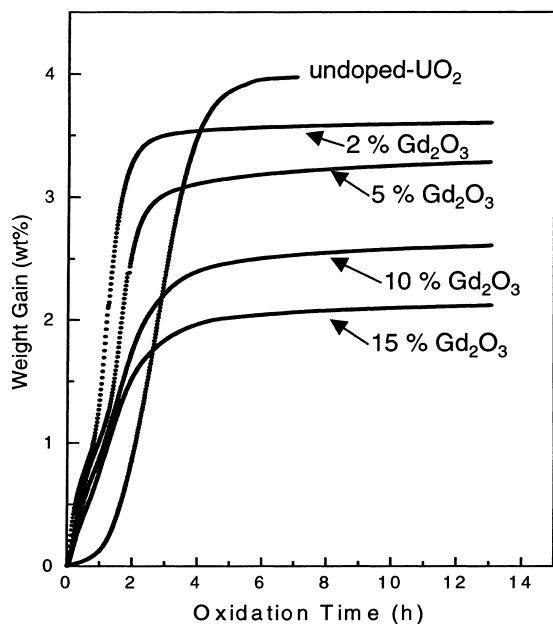


Fig. 2. Oxidation behavior of Gd-doped UO_2 at 350°C .

3.3. Irradiated- UO_2 fuels

3.3.1. Gd-doped fuels

Irradiated-Gd-doped UO_2 specimens with 10 and 28 GWd/tU burnups were oxidized at $275\text{--}400^\circ\text{C}$ in the unlimited air environment and the results are shown in Fig. 4. The rates of oxidation in all samples are very fast at first, and then slowly increase up to about 3% weight gain. This figure also shows that the oxidation rates in the same burnup samples increase with test temperatures. However, at 275°C the specimen with 10 GWd/tU burnup shows a little higher oxidation rate than that of the specimen with 28 GWd/tU. Fig. 5 shows the comparison between irradiated (bold curve) and unirradiated (dotted curve) Gd-doped UO_2 oxidation results at 350°C . Comparing the irradiated-6% Gd-doped fuel to the unirradiated-5% Gd-doped fuel, the oxidation rate of the irradiated one is much lower than that of the unirradiated one. This means that the oxidation of irradiated-Gd-doped fuel depends on both the dopant and irradiation.

3.3.2. Gd-undoped fuel

The oxidation behavior of irradiated- UO_2 at $275\text{--}400^\circ\text{C}$ is shown in Fig. 6. For comparison, the $300\text{--}375^\circ\text{C}$ oxidation results of SIMFUEL SFOs having no simulated fission products are also plotted. The burnup levels of irradiated- UO_2 fuels were between 16 and 38 GWd/tU. Numerical values in parenthesis in this figure indicate the burnup levels in terms of GWd/tU. As shown in this figure, the unirradiated fuels (SIM-

FUEL SFOs) are oxidized so fast that the weight gains at all temperature regions reach the saturation levels before 50-h oxidation time. However, the irradiated samples show relatively slow oxidation rates and a trend of lower intermediate-levels of weight gain than those of unirradiated ones. The intermediate-levels of weight gain depend on test temperatures. At 350°C and 400°C , however, these samples indicate nearly the same oxidation rates compared to unirradiated- UO_2 and the saturation levels of irradiated- UO_2 are a little higher than those of unirradiated ones. It is also revealed that the different burnup specimens oxidized at the same temperature show different oxidation kinetics. At 275°C , the 38 GWd/tU burnup specimen shows an intermediate-level of weight gain of about 2.5 wt% after 300 h oxidation. In contrast, the 16 GWd/tU specimen shows a higher level of weight gain and a transition phenomenon. This specimen shows a typical oxidation behavior of irradiated UO_2 until 140-h oxidation time (2.5% weight gain point), and then a rapid re-increase phenomenon accompanying localized powdering. The oxidation proceeded to obtain about 4% weight gain. This kind of transition also appeared on 38 GWd/tU burnup specimens at 300°C and 330°C temperatures.

4. Discussion

4.1. Additive effects

McEachern and Taylor [10, and references therein] pointed out that the observed correlation between the t_p and burnup is likely related to the large amount of fission-product elements in high-burnup fuel. Choi et al. [12] also confirmed these facts by using simulated fuels.

In the present research, however, we focus on studying the kinetic changes by some additives, so the increase patterns of weight gain rather than phase changes were mainly observed. Before the experiment it is already expected that these oxidation patterns on SIMFUEL cannot simulate exactly the kinetic behavior of irradiated UO_2 . Because SIMFUELS can simulate only the effect due to fission fragment elements formed in irradiated UO_2 . But there are some other factors affecting the oxidation patterns of irradiated UO_2 . One of them is the effect of fission gas bubbles forming along the grain boundary during irradiation of UO_2 . It has been known that these bubbles may cause the rapid diffusion of oxygen along grain boundaries and so the fast oxidation of irradiated UO_2 can occur at an early stage.

In the present experiments, it is found that SIMFUELS containing no fission gas bubbles in the grain boundaries, do not show fast oxidation rates at the

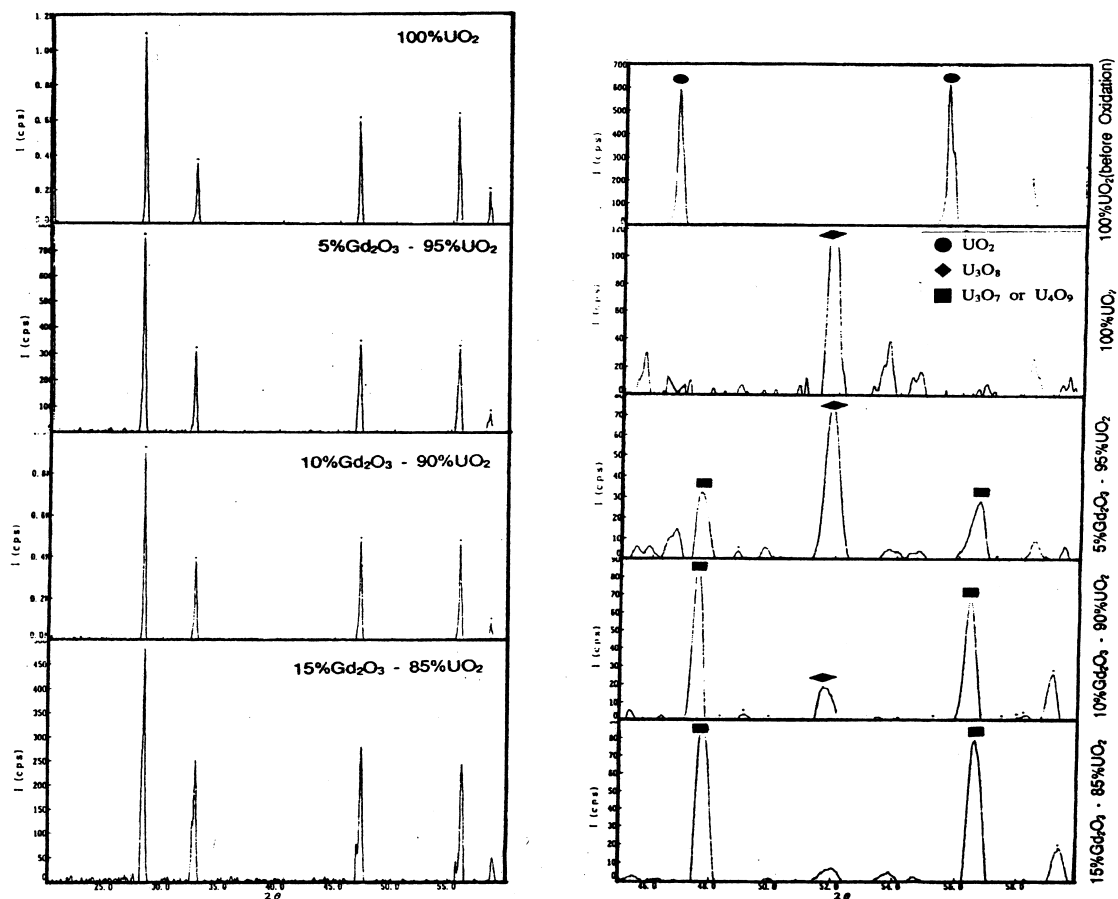


Fig. 3. (a) X-ray diffraction traces for Gd-doped UO_2 before oxidation. (b) X-ray diffraction traces for Gd-doped UO_2 oxidized in air at 350°C for 15 h.

initial stages. While continuing oxidation they only show the delaying features of an increase rate of oxidation weight gain. They do not show the long-time sustaining features at intermediate-levels of weight gain as irradiated UO_2 usually indicates.

In the oxidation mechanistic study of UO_2 with doping materials, such as gadolinia, McEachern and Taylor [10] reviewed that the oxidation stability of the cubic fluorite phase in doped UO_2 enhanced relative to the undoped material, so that there is a qualitative change in the nature of the oxidation mechanism; i.e. the intermediate U_4O_{9+y} is observed rather than U_3O_7 . Thomas et al. [13] and Cambell et al. [14] oxidized UO_2 and gadolinium-doped UO_2 samples at low ($175\text{--}200^\circ\text{C}$) temperatures. XRD patterns of partially oxidized samples indicated that oxidation of pure UO_2 (or UO_2 doped with small amounts of impurities) undergoes via U_3O_7 , whereas UO_2 doped with large amounts (4–10 wt%) of Gd_2O_3 is oxidized via U_4O_{9+y} . Many other investigators [10, references therein] have also studied the doping effects on UO_2 oxidation.

Our present results from thermal and XRD tests on Gd-doped UO_2 in Figs. 2 and 3 can also support the above mentioned mechanistic analyses, but in view of kinetic pattern changes it is found that the doped fuels are oxidized faster than the undoped ones at the initial stage of oxidation. This fact implies that an additive with a certain quantity may affect the increase of the initial oxidation rate as well as the sustenance of intermediate-levels of weight gain at the later stage. From this fact, it might be possible to guess that the fission elements in irradiated UO_2 can also affect the initial kinetic behavior.

4.2. Irradiation effects

The various oxidation data of irradiated UO_2 reported from several laboratories show quite different kinetic behaviors, and sometimes they show the opposite results. Boase and Vandergraaf [15] found that powdered and irradiated CANDU fuels were oxidized 3–3.5 times faster than unirradiated UO_2 powders between

200°C and 300°C; however, they observed no significant differences in the rates of oxidation between UO_2 pellets and irradiated-fuel fragments at 320–460°C. Similarly,

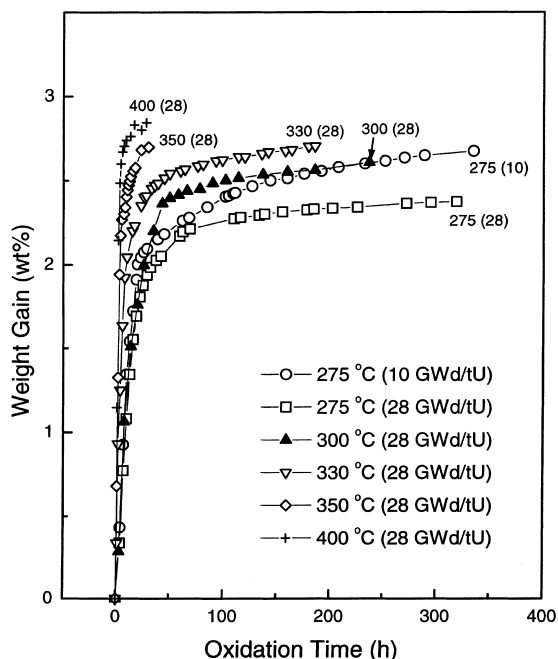


Fig. 4. Oxidation behavior of irradiated-Gd-doped UO_2 .

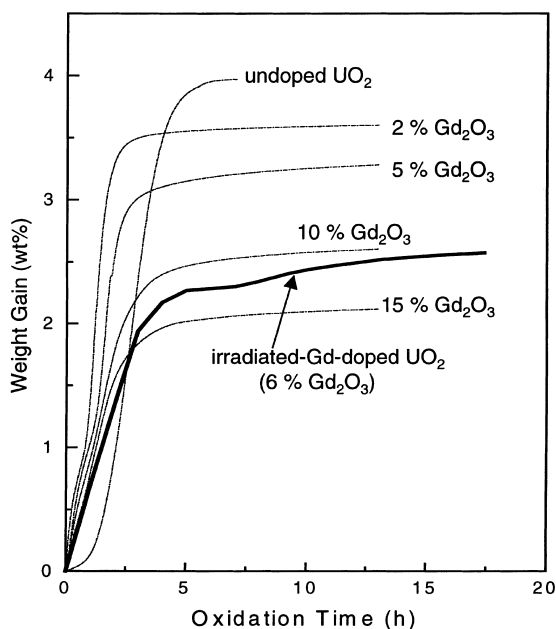


Fig. 5. Comparison of oxidation results between irradiated and unirradiated-Gd-doped UO_2 at 350°C.

Hastings et al. [16] reported that irradiated CANDU fuel is oxidized 2–3 times faster than unirradiated UO_2 at temperatures below 300°C, but that the oxidation occurs at similar rates in the range of 300–400°C. Schmets [17] found that with bulk specimen of irradiated UO_2 , the oxidation rates are higher than those of unirradiated UO_2 samples.

However, contrary to the above results Olsen [18,19] reported that diametric enlargement of the BWR type rod (because of U_3O_8 powder formation) occurs most quickly at the ends of the rod (which have the lowest burnup). Harrison et al. [20], using irradiated 120 μm -diameter UO_2 spheres, also found oxidation rates are lower than those of unirradiated ones. Gilbert et al. [21] showed that unirradiated- UO_2 or irradiated- UO_2 fuel with a burnup below 15 GWd/tU is oxidized to U_3O_8 more readily than irradiated- UO_2 fuel with a burnup greater than 15 GWd/tU . Campbell et al. [22] also reported that oxidation behavior of LWR spent fuel differs markedly from unirradiated UO_2 pellets. Compared with unirradiated fuel pellets, initial rates of weight gain of PWR spent fuel are up to 50 times greater, the period for powder to form is longer, and powder formation begins at higher weight gain. Recently, Kim et al. [23] have also confirmed that the rate of U_3O_8 formation with high-burnup UO_2 (37 GWd/tU) and Gd-doped UO_2 (10 and 27 GWd/tU) fuel at 275°C is much slower than that of an unirradiated UO_2 pellet. McEachern and Taylor also presented in their reviewing paper conclusively [10] that the rate of $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ formation in spent fuel is between 2 and 50 times faster than unirradiated UO_2 .

From the above opposite results, although the reasons of oxidation kinetic differences between both types of fuels are not clear, it may be supposed that the oxidation rate differences are due to fuel types (CANDU and BWR/PWR) and burnup levels. Our present experimental results using PWR type fuel (as shown in Fig. 6)

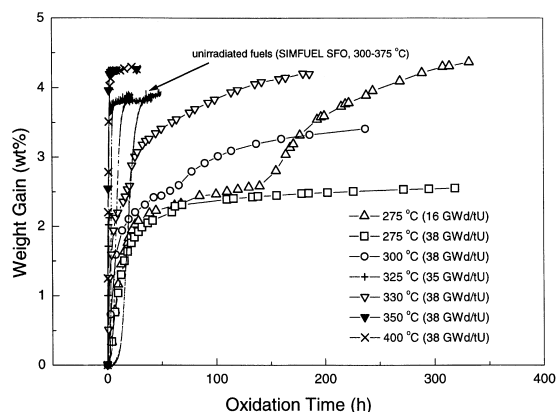


Fig. 6. Oxidation behavior of irradiated and unirradiated UO_2 .

are comparable to those of the above mentioned BWR/PWR cases and in relatively low temperature ranges it is found that irradiated samples have more resistive properties against oxidation than unirradiated ones do. Our present results and the above cited ones cannot distinguish clearly between the two transitions, UO_2 to $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ and $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ to U_3O_8 , as described and defined in [10]. If the phase changes are confirmed, it is possible to explain that for irradiated fuels the first transition (UO_2 to $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$) is quite rapid and the second one ($\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ to U_3O_8) is much slower at relatively low oxidation temperatures compared to unirradiated fuel.

Another interesting fact we found is a transition phenomenon of relatively lower burnup fuels in the mid-range of weight gain, as shown in the 16 GWd/tU burnup sample at 275°C in Fig. 6. Higher burnup fuels (as shown in 38 GWd/tU burnup sample at 275°C in Fig. 6) do not show this kind of transition, but showed to sustain an intermediate-level of weight gain. This fact means that there is a certain minimum burnup needed for obtaining stabilization effect by irradiation against continuing oxidation, such as powdering or U_3O_8 formation. Our former experiments reported in [23] also indicate similar results comparable to those of present experiments.

As shown in Fig. 7, it was also found that the oxidation behaviors between irradiated-Gd-doped and -undoped fuels showed big differences. Gd-doped ones are more resistive than undoped ones and they did not show the kinetic transition phenomenon as undoped ones do.

Summing up the above results, it may be concluded that the positive correlation between t_p and burnup for PWR spent fuel is valid over a certain burnup level.

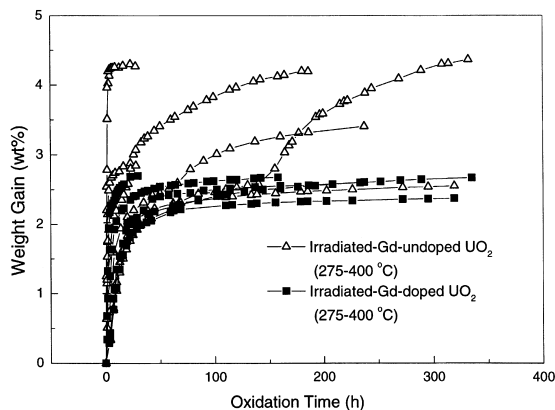


Fig. 7. Comparison of oxidation behaviors between irradiated-Gd-doped and -undoped UO_2 .

4.3. Comparison between kinetic changes by additive addition and irradiation

Irradiation effects on UO_2 oxidation are very similar to additive effects on fuels with doping elements, such as SIMFUEL and Gd-doped fuels.

In the case of irradiated fuels, it is revealed that the oxidation rate in the initial stage is very fast and stabilized at an intermediate-level of weight gain. In addition, transition phenomenon at a certain reaction point appears very clearly in the case of lower burnup samples. In unirradiated-Gd-doped UO_2 the oxidation rate appears to be comparatively fast at first, and the intermediate-level of weight gain is lower than those of Gd-undoped UO_2 and decreases with the increase of Gd content. Also 2 and 5% Gd-doped UO_2 at 350°C show the transition phenomenon at 1–1.5 wt% point, as shown in Fig. 8. Therefore, it can be suggested that the Gd-doped fuels indicate very similar aspects to that of irradiated fuels.

In the case of SIMFUEL, oxidation rate shows a typical sigmoidal curve like that of unirradiated fuel and the oxidation rates are delayed as the simulated burnup levels increase. This delaying rate also shows on irradiated fuel. However, the saturation levels in all SIMFUEL samples appear to reach a theoretical level when complete oxidation occurs. Conclusively, the oxidation behavior of unirradiated-Gd-doped UO_2 is more similar to that of irradiated- and -Gd-doped UO_2 than that of SIMFUEL.

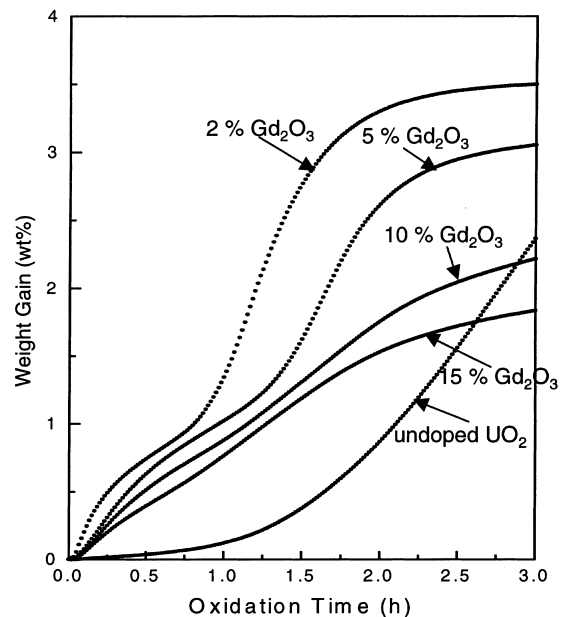


Fig. 8. Oxidation transition behavior of Gd-doped UO_2 at 350°C.

It has been well known that the initial oxidation in irradiated UO_2 is mainly the formation of $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ [10]. It has been explained that the rate of $\text{U}_3\text{O}_7/\text{U}_4\text{O}_9$ formation in spent LWR fuel is rapid, relative to that of unirradiated UO_2 , partly because of the concentration of fission gas bubbles along grain boundaries [24]. However, it has not yet been explained how much the fission gas bubbles in grain boundaries can affect the initial oxidation of irradiated fuel quantitatively and needs further investigation. In the present experiment, it is found that unirradiated-Gd-doped UO_2 having no grain boundary bubbles also indicates a faster oxidation rate at the early stage than that of the Gd-undoped UO_2 . This fact may imply that the doping elements, such as Gd, can also affect the initial oxidation kinetics of UO_2 , and so it can be suggested that the fission element materials formed in irradiated UO_2 should be also one of the reasons for the initial fast oxidation behavior of irradiated- UO_2 .

5. Conclusions

The changes of oxidation kinetic behaviors of UO_2 by an additive addition and irradiation in air have been investigated.

At 300–375°C, oxidation rates of SIMFUEL show a typical sigmoidal curve and decrease with the simulated burnup increase, but the saturation levels are almost identical.

It is also revealed that the intermediate-levels of weight gain of oxidized unirradiated-Gd-doped UO_2 are much lower than those of undoped UO_2 , and the levels decrease with the content of Gd. The initial weight increases of all Gd-doped UO_2 samples are faster than that of undoped- UO_2 2% and 5% Gd-doped UO_2 show the transition phenomenon at a certain oxidation point. The XRD results of Gd-doped UO_2 reveal that 15% dopant levels can inhibit the formation of U_3O_8 on the surface of UO_2 .

In the case of irradiated-Gd-doped UO_2 , the patterns of oxidation curves are similar to unirradiated-Gd-doped UO_2 , and the intermediate-levels of weight gain increase with the temperature, but they do not exceed 3 wt%. The specimen with 10 GWd/tU burnup show the oxidation rate to be a little higher than the specimen with 28 GWd/tU at 275°C.

In the experiments of irradiated-Gd-undoped UO_2 , compared to the results of unirradiated UO_2 samples at 300–375°C, the irradiated ones are oxidized faster than unirradiated ones and indicate sigmoidal kinetic curves and a same saturation level. Clear transition phenomenon of oxidation rates also appeared in the irradiated UO_2 with 16 GWd/tU burnup at 275°C and with 38 GWd/tU burnup at 300°C and 330°C.

Acknowledgements

This study has been carried out under the Nuclear R&D Program by MOST (Ministry of Science and Technology) in Korea.

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