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RE-ASSESSMENT OF NITRIDE FUEL POTENTIAL IN THE CURRENT CONTEXT OF THE NUCLEAR INDUSTRY

Sylvie M. Reynaud and Kenneth L. Peddicord
Texas A&M University
Department of Nuclear Engineering
M.S. 3133
College Station, Texas 77843-3133
E-mail : sylvie@tamu.edu, k-peddicord@tamu.edu

ABSTRACT

In recent years, there has been a renewal of interest in nitride fuels throughout the international community. The new challenges met by the nuclear industry, which include greater safety margin and the question of the waste management, have resulted in active research programs in next generation fast spectrum reactors and waste transmutation systems. Through these programs, nitride fuel has emerged as one of the most promising advanced fuels, thanks to their numerous favorable properties.

INTRODUCTION

The concept of nitride fuels is not a new one. It was first investigated during the 60s during the development of the liquid metal fast breeder reactors (LMFBRs) as an alternative to oxide and metal fuels. In the U.S., the first systematic investigation started with the Advanced LMFBR Fuels Development Program, which began in 1974 [1]. The results were promising but, because of the parasitic neutron absorption by N-14 in nitride fuel, the carbide fuel system was preferred for further explorations. At that time, the principal objective of the fast reactor program was to have the highest breeding gain possible and such neutron absorption with a nitride system was a serious disadvantage. Then, with the progressive decrease in interest in LMFBRs, the development of these advanced fuels gradually ceased, if not totally abandoned. However, the changes encountered by the nuclear industry during the past two decades have renewed the interest in the nitride systems. The first indication of the return of nitride fuels was the choice of uranium mononitride as fuel for the SP-100 project in 1984 [2]. Today, numerous research projects all around the world, dealing directly with the use of nitride fuels, are a sure indication of this renewed interest.

Among the new challenges encountered by the nuclear industry, the question of greater safety margins and the problem of the waste management occupy a large part. In the current context, a new generation of fast reactors, with enhanced performance and safety, offer a solution for a long-term energy source and the transmutation of the nuclear waste may offer a solution for the waste question, which is central for the public. Research programs have shown that nitride fuels have a large role to play in the new reactor generation, thanks to their numerous advantageous properties.

SUMMARY OF NITRIDE FUEL PROPERTIES

The general properties of nitride fuels were initially collected during the investigation phase of the fast reactor fuel program and they have been summarized, as well as the potential of this kind of fuel, in the early 70's [3].

The dominant characteristic of nitride fuels is that they combine properties of both metallic and oxide fuels. They present a high heavy metal density and a very good thermal conductivity, features generally associated with metal fuels. On the other hand, they present a favorable Doppler coefficient, as oxide fuels. Table 1 gives a comparison of different properties for metal, oxide and nitride fuels.

Table 1: Comparison between metal, oxide and nitride fuels properties [5]

	Metal		Oxide		Nitride	
	U	Pu	UO2	PuO2	UN	PuN
Melting point (°C)	1132	640	2730	2300	2600	2500
Theoretical density (g/cm^3)	19.05	19.86	10.96	11.46	14.32	14.22
Heavy atom density (g/cm^3)	19.05	19.86	9.66	10.11	13.51	13.43
Heat conductivity at 500°C (W/cm°C)	0.3		0.047		0.12	
Specific heat (cal/g.°C) at 1500°C (metal at 500°C)	0.040	0.044	0.081	0.084	0.065	0.061

Another interesting point is the very good compatibility of nitride fuels with the liquid metal coolants and the cladding materials. For example, this compatibility with the coolant allows sodium bounding when sodium is used as the coolant,, which increase the thermal performance of the fuel pin. This compatibility also reduces the risk of accident when cladding failure occurs. As far as the cladding is concerned, the chemical interaction between fuel and cladding can even be neglected [4].

APPLICATIONS IN NEXT GENERATION REACTORS

As mentioned earlier, the new applications for nitride fuels are the enhancement of plant safety and the transmutation program. Since the focus of this paper is for civil reactors, the application of nitride fuels for space power will not be presented. However, the experience gained in this program has also been helpful for the development of the others by, for instance, giving new irradiation data.

Advanced Fast Reactors:

At the beginning of the liquid metal fast breeder reactor development, the main objective was to achieve a breeding ratio as high as possible with the shortest doubling time. Today, priorities have changed, and the enhancement of performance and safety is the order word. To achieve these goals, the choice of the fuel is important. Nitride fuels, thanks to their favorable properties, offer the possibility of better performance and enhanced safety. Lyons et al. [4] summarized these effects of the nitride fuel characteristics on the reactor system, as shown in Table 2.

These positive aspects have been confirmed by calculations and experiments for different situations (power levels, coolants) [6;7]. Other programs are acquiring data to demonstrate the better behavior of nitride fuels in comparison to traditional oxide fuels, particularly in the field of passive safety [8].

As explained earlier, the concept of nitride fuels is not a new one. The possible advantages offered by this kind of fuel have long been recognized. However, some drawbacks were first encountered, which limited then their potential. Today, these drawbacks have been overcome, which permits the new interest. The two main drawbacks were:

- The dissociation at high temperature
- The (n,p) reaction with N-14

At high temperatures, a dissociation of the fuel can occur: free uranium and N_2 gas are formed, even below the fuel melting point. This problem has been solved by a strict control of the stoichiometry during the fabrication of the fuel, and imposing a design limit temperature for security margin. This control does not represent anymore a challenge for the current fuel fabrication techniques. Recent studies [9] have also shown than the thermal decomposition of nitride fuel does happen for temperatures at least 1800 °C for various atmospheres, which give a large operating temperature margin.

The ^{14}N, which constitutes the main part of nitrogen in the nitride fuel, exhibits a rather large cross section for the (n,p) reaction giving ^{14}C. This phenomenon has two consequences: the formation of radiotoxic ^{14}C and the decrease of the breeding gain due to the loss of neutrons. Concerning this reaction involving ^{14}N, two positions exist currently: (1) to consider that this problem and it needs to be answered or, on the other side, (2) to consider that this is not a problem anymore in view of the current objectives of the advanced reactors. This second position is based on the fact that the objective of the next generation of fast reactors is not to have the highest breeding ratio as possible. For some design propositions, like long-life reactors, a breeding gain superior to unity is still required, but the purpose is not to produce plutonium anymore and a breeding gain slightly superior to one is sufficient. Therefore, the parasitic neutron absorption by ^{14}N is no longer considered as a problem. However, even in this context, some concerns have aroused about the ^{14}C production, which might harm the environment and complicate eventual reprocessing [10]. The solution to this problem is to enrich the fuel with 90-99 % of ^{15}N. Several methods are available to perform this enrichment: low temperature rectification, chemical exchange, gas diffusion and centrifugation, or use of electrical discharge or laser beam. The challenge is now to be able to realize this isotopic enrichment without dramatically increasing the total cost of the fuel manufacturing.

Table 2: Nitride Fuels Characteristics (Lyons et al., 1991)

Characteristic	System Impact
High thermal conductivity and heavy metal density	High power density, high thermal performance, good core breeding, low reactivity swings, and long residency time compared to oxide fuels
Fuel, cladding, and coolant compatibility	Post-breach operation possible, minimal fuel/cladding chemical interaction, allows use of sodium bond.
Low sodium void coefficient and favorable Doppler coefficient	Enhanced safety performance due to higher margins to failure, excellent tolerance for loss of flow and transient overpower without scram as compared to metal fuels
Low fission gas release and fuel swelling	Short plenum and pins, low cladding stresses, longer pin lifetime, and better economics

Waste Transmutation:

Another field of research responsible for the renewal of interest in nitride fuel is the development of the waste transmutation. The international community is actively pursuing research to find the optimum fuel system. The concept of transmutation does not only involve the burning of the actinides, but it also includes the total fuel cycle, particularly the recycling process through which the actinides would be separated. Therefore, the chosen fuel must not only present good in pile performance, including enhanced safety features, but also allow easy recycling.

Nitride fuels offer the possibility to use already existing recycling process (PUREX process and pyrochemical process). Many investigations have been undertaken in order to exploit to a maximum this recycling properties of the nitrides and to create a concept of recycling process for actinide burning. Several options have been presented, for instance the Japanese “LINEX” process [11]. A complete scheme, involving standard PWR commercial reactors and new advanced actinide burner fast reactors, is in planning [12].

The compatibility of nitride fuel with the recycling process (in contrary to carbide fuel for instance) is not the only advantage that they present for waste transmutation. Its thermal and neutronic properties allow the burning necessary for transmuting the minor actinides. In addition, as previously mentioned, the use of nitride fuel can enhance the safety of the transmuter. Specific safety studies were performed for a core dedicated to waste transmutation, taking into account the particularity of the fuel (with high content of minor actinides), and they demonstrate the very good behavior of a nitride fueled core in term of safety [13].

CONCLUSION

The new imperatives that are currently driving the nuclear industry have given a second wind to the fast reactor applications. They have meanwhile renewed the research for advanced fuels that would fulfill their need. Nitride fuels, thanks to their intrinsic properties, appear as a front line candidate, especially in application for the waste transmutation. However, only few experimental data are currently available on nitride fuels and more are needed to pursue it any further. That is why an aggressive irradiation program has been initiated worldwide.

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