

การพัฒนาเชื้อเพลิงสำหรับโรงไฟฟ้านิวเคลียร์แบบเครื่องปฏิกรณ์น้ำมวลเบาและ เครื่องปฏิกรณ์ปรมาณูวิจัยแบบ TRIGA

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บทคัดย่อ

ได้ทำการสร้างแท่งเชื้อเพลิงจำลองสำหรับเครื่องปฏิกรณ์ปรมาณูวิจัยแบบ TRIGA โดยมีขนาดและลักษณะภายนอกคล้ายคลึงกับของจริงโดยใช้วัสดุเหล็กกล้าไร้สนิม ได้ทำการสร้างแท่งเชื้อเพลิง TRIGA 2 แท่งที่บรรจุโลหะเหลวภายในร่วมกับองค์ประกอบภายในจำลอง หลังจากตัดเพื่อให้เห็นองค์ประกอบภายในพบว่าโลหะเหลวที่อยู่ภายในมีความครบถ้วนสมบูรณ์และไม่เกิดช่องว่างขึ้นในโลหะเหลว ได้ทดสอบ material compatibility โดยจุ่มแผ่นเหล็กกล้าไร้สนิมหลายแผ่นลงในโลหะเหลวที่บรรจุอยู่ใน 2 ภาชนะบรรจุที่ 120°C และ 250°C ซึ่งหลังจากผ่านมาเป็นเวลา 1 ปี 8 เดือน ไม่พบการกัดกร่อนบนแผ่นเหล็ก ได้ทำการผลิตเม็ดเชื้อเพลิงนิวเคลียร์สำหรับโรงไฟฟ้านิวเคลียร์แบบเครื่องปฏิกรณ์น้ำมวลเบา (light water reactor) โดยใช้ผงยูเรเนียมออกไซด์ธรรมชาติที่สกัดจากสินแร่โมนาไซด์ผสมกับตัวประสานที่เหมาะสม และอัดให้เป็นเม็ดทรงกระบอกตันขนาดเส้นผ่านศูนย์กลางประมาณ 1.2 ซม. ในโมลด์และ puncher ที่ทำจาก tungsten carbide ในเครื่องอัดไฮดรอลิกส์ขนาดแรงอัด 100 ตัน หลังจากนั้นทำการเผาประสานที่อุณหภูมิ 1200°C ในบรรยากาศเฉื่อยเป็นเวลา 72 ชั่วโมง ผลที่ได้คือเม็ดเชื้อเพลิงมีความหนาแน่นสูงถึง 85.9 % TD

คำสำคัญ : เชื้อเพลิงนิวเคลียร์ ยูเรเนียม โรงไฟฟ้านิวเคลียร์ เครื่องปฏิกรณ์ปรมาณูวิจัย

Development of Fuels for Light Water Reactor Nuclear Power Plant and TRIGA Research Reactor

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Abstract

A mock up of the TRIGA research reactor fuel rod was fabricated from stainless steel with the size and appearances similar to the actual fuel rod. Two TRIGA fuel rods were fabricated, being loaded with liquid metal and simulated internal components. After the rods were cut to observe inside, liquid metal was found to be very intact without any void formation. Material compatibility test was performed by immersing several stainless steel plates in two containers containing liquid metal at 120°C and 250°C. After 1 year and 8 months, no corrosion has been observed. Nuclear fuel for light water reactor nuclear power plant was fabricated. Natural uranium oxide powder extracted from monazite ore was mixed with an appropriate binder and compressed into a solid cylinder ~ 1.2 cm. in diameter inside a 100-ton hydraulic press using a mould and a puncher made of tungsten carbide. Subsequently, the pellet was sintered at 1200°C in an inert atmosphere for 72 hours. The achieved sintered pellet density was 85.9 % TD.

Keywords: nuclear fuel, uranium, nuclear power plant, research reactor

1. Introduction

Conventional TRIGA fuel rod composes of uranium-zirconium hydride (UZrH) fuel meats inside a stainless steel cladding. To illustrate that Thailand has a capability to fabricate the cladding tube, a mock up of the TRIGA fuel rod was fabricated from stainless steel with the size and appearances similar to the actual fuel rod.

In an as-fabricated TRIGA fuel rod, there is an air-filled gap between the fuel meat and the cladding. To help improve heat transfer across the gap, a liquid metal (LM) compound composing of 33 wt.% each of Pb, Sn and Bi was proposed to fill in the gap. This is a non-reactive and low-melting-point alloy which melts at ~ 96°C. The presence of LM in the gap would help make the fuel safer because of the reduced operating temperature of the fuel meat and would potentially allow the fuel to operate at higher burnups because the gap can be made large enough to accommodate fuel swelling during the entire service life without compromising the fuel operating temperature. Liquid metal in the gap can also limit interactions between the fuel and steam by impeding steam ingress into the gap following cladding failure. These similar benefits were also realized in research works that studied replacing helium in the fuel-cladding gap of LWR fuel rod with LM of the same composition^{1,2,3}. In addition, for LM-bonded LWR fuel rods, massive secondary hydriding can also be prevented.

Due to a surface tension effect, voids or unfilled spaces in the liquid metal may form when

the metal fills in a small gap. Were this to occur, it could lead to local-overheating of the fuel meat next to the void. Experiments were made to confirm that the fuel rods were void-free.

Furthermore, with the service lifetime of several years and with the normal operating temperature of about 120°C, LM may (or may not) corrode the stainless steel cladding. Thus, another experiment was carried out to study the compatibility of LM and stainless steel at normal operating temperature and elevated temperature for an extended period of time.

Since Thailand may consider building nuclear power plants in the future and since Thailand Institute of Nuclear Technology (TINT) realizes that the country may have to one day fabricate the fuel pellets by herself, studies were made to fabricate nuclear fuel pellets for light water reactors. The raw material was natural uranium oxide powder extracted from monazite ore. The goal was to fabricate LWR fuel pellets with natural ^{235}U enrichment exhibiting the physical appearance and density close to that of commercial UO_2 fuel pellets.

2. Materials and Equipment

2.1 Development of fuels for TRIGA research reactor

304-grade stainless steel tubes, bars, sheets and containers, brass bars, Pb-Sn-Bi compound (33 wt.% each, at least 99% purity and different elements purchased from different suppliers), digital temperature controllers (assembled from several parts), K-type thermocouples (Omega brand), heating tapes (Brisk Heat brand), insulator, aluminum foil.

2.2 Development of fuels for light water reactor

Uranium oxide powder (natural ^{235}U enrichment extracted from monazite ore provided by TINT), hydraulic press (TMC brand, 100 tons capacity), stearic acid, tungsten carbide mould and puncher, ball mill roll, high-temperature horizontal furnace (assembled from several parts), research-grade Ar gas (Praxair brand), gas flow rotameters, optical microscope.

3. Methodology

To fabricate a mock up of the TRIGA fuel rod, outer dimensions of the TRIGA fuel rod displayed at TINT were measured and the fuel rod were fabricated conforming to such dimensions.

To make certain that voids did not exist, two mock up TRIGA fuel rods were fabricated from stainless steel tubes with dimensions similar to that of the actual fuel rod. They were loaded

with simulated fuel meats, simulated graphite reflectors and LM in the amount sufficient to fill in the entire gap. The rods were cut to observe inside.

To study the compatibility of LM and stainless steel, a compatibility test was setup. It consisted of two stainless steel containers each filled with LM to about $\frac{1}{2}$ of the capacity. Each container was surrounded by a heating tape and further wrapped with an insulator and an aluminum foil. A type-K thermocouple was immersed into the LM in each container. The heating tape and the thermocouple were connected to a digital temperature controller. Six stainless steel bars were immersed into the LM in each container. The temperature in the first container was maintained at 120°C to simulate the normal operating condition while that in the second container was maintained at 250°C to allow observation of any corrosion in an accelerated manner. Each stainless steel bar was removed from the container after a certain amount of time has elapsed to observe any corrosion.

To develop fuel pellets for light water reactor, an appropriate amount of UO_3 powder was heated at about 800°C in argon for about 8 hours inside a high-temperature furnace in order to turn it into a black U_3O_8 powder. The UO_3 powder was supposed to be heated in a reducing atmosphere at $\geq 1500^{\circ}\text{C}$ for hours to completely reduce to UO_2 . However, in this phase of the study, it was deemed too difficult to do so because several parameters needed to be correct to fully reduce to uranium dioxide. Therefore, U_3O_8 powder was chosen as the starting material for pellet fabrication.

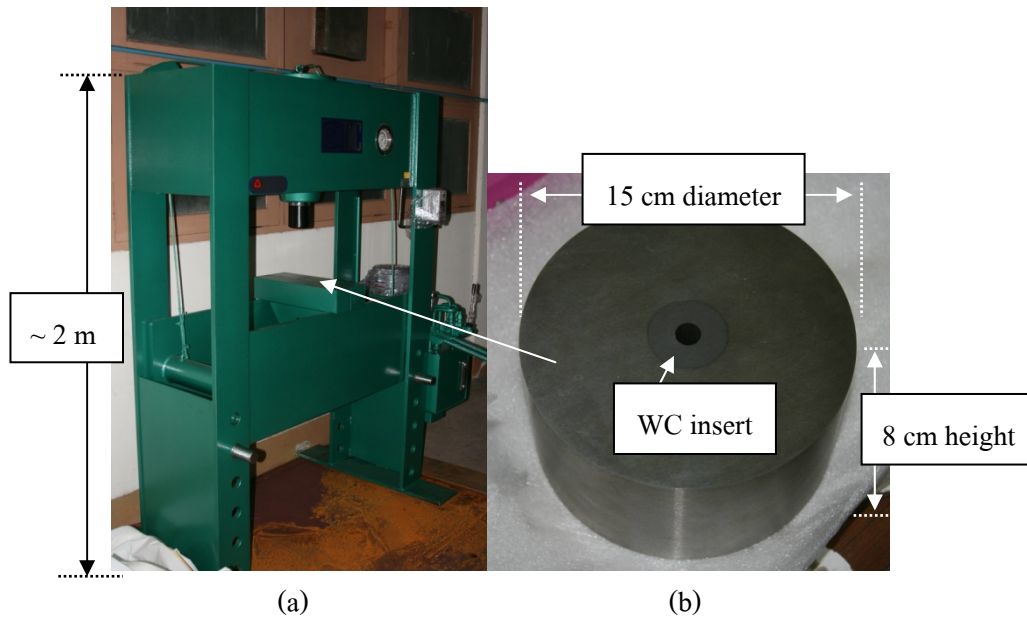


Fig. 1 (a) 100-ton hydraulic press machine and (b) the mould (made of tungsten carbide insert into Cr12 steel body with HRC 58)

The resulting U_3O_8 powder was homogeneously mixed with a few percent of stearic acid as a binder in a ball mill roll. The mixture was then poured into a tungsten carbide mould with a 1.2-cm diameter hole and was subsequently compressed by a tungsten carbide puncher in a 100-ton hydraulic press machine. The pellet was removed from the mould and sintered at 1200°C in a pure argon atmosphere for 3 days inside the high-temperature furnace. Fig. 1 shows the 100-ton hydraulic press machine and the mould.

4. Results and Discussions

Fig. 2 illustrates the fabricated mock up of the TRIGA fuel rod. The rod was successfully fabricated with fine details very similar to those of the actual fuel rod.



Fig. 2 Successfully fabricated mock up of the TRIGA research reactor fuel rod

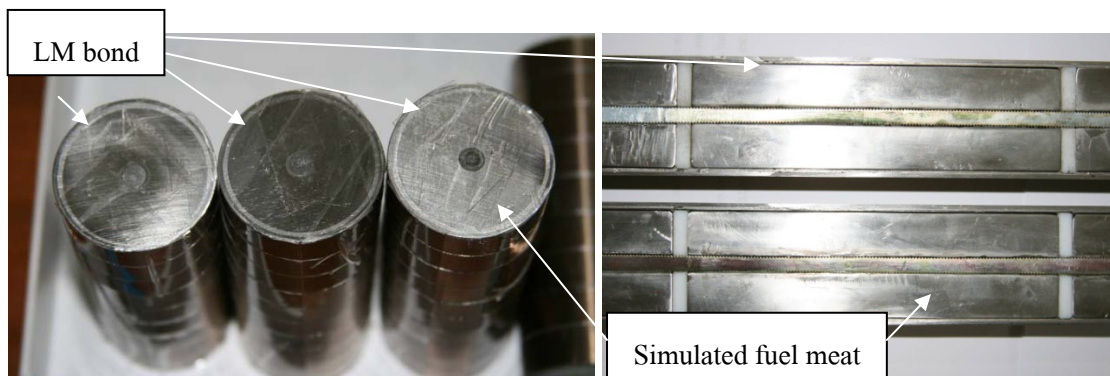


Fig. 3 Simulated TRIGA fuel rods, which had been cut to observe the LM inside.

Fig. 3 displays simulated TRIGA fuel rods loaded with simulated fuel meats, simulated graphite reflectors and LM, which had been cut to observe inside. It was evident that there was no observable void formation in the LM bond. Formation of fission gas bubbles in the LM during

irradiation can be neglected because of the very low fission gas release from hydride fuel (release fraction $\sim 10^{-4}$ up to 600°C)⁴.

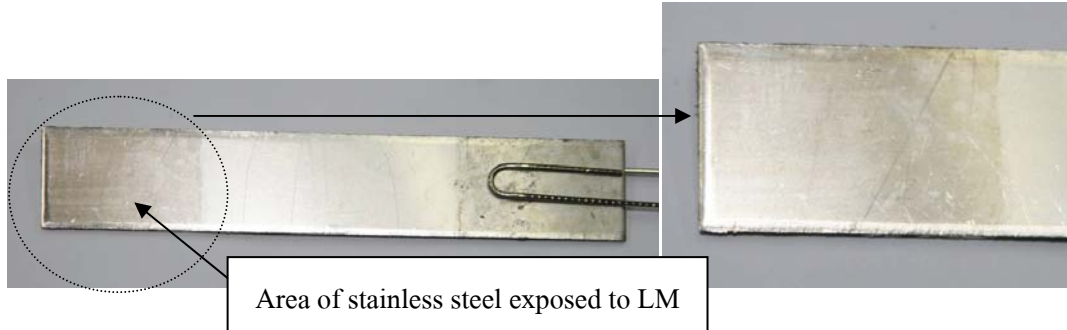


Fig. 4 Example of stainless steel plate used in the compatibility study.

Fig. 4 illustrates an example of the stainless steel plate used in the compatibility study. It was clearly seen that there was no observable corrosion, even at after 1 year and 8 months at 250°C . Thus, from the materials compatibility point of view, LM can be safely used to fill in the gap.



Fig. 5 Uranium oxide pellet after it was compressed (left) and sintered (three on the right).

Fig. 5 displays the uranium oxide pellet after it was compressed and sintered. It can be seen that the compressed pellet exhibited a very satisfactory feature as the surface was very smooth and had a shiny dark color. After sintered, however, there were some cracks appearing on the pellet surface. But, overall, it exhibited a physical appearance similar to that of the actual UO_2 pellet. The density of the sintered pellet was 85.9% of the theoretical density of U_3O_8 . This was considered a successful result overall. Work in the next phase includes turning UO_3 into UO_2 and producing crack-free pellets with the density close to 96 %TD⁵ of UO_2 , the density of LWR pellets. Factors to be studied to reduce UO_3 into UO_2 include hydrogen concentration in the flow gas, temperature and time. Factors to be studied on sintering UO_2 pellets include flow gas composition, temperature, time and percentage of the binding agent.

5. Summary

A mock up of the TRIGA research reactor fuel rod was successfully fabricated. LM was found to completely fill in the fuel meat-cladding gap with no observable void formation. No corrosion had been observed between stainless steel bars and LM even at 250°C for 1 year and 8 months. Sintered U_3O_8 pellet exhibited a physical appearance similar to that of the actual UO_2 pellet, albeit some cracks formed on the surface after sintering. The achieved sintered pellet density was 85.9 % TD. The goal for the next phase of the study was to sinter UO_2 pellets to 96 % TD.

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