



## Original Article

## Uranium thermochemical cycle used for hydrogen production

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## ABSTRACT

Thermochemical cycles have been predominantly used for energy transformation from heat to stored chemical free energy in the form of hydrogen. The thermochemical cycle based on uranium (UTC), proposed by Oak Ridge National Laboratory, has been considered as a better alternative compared to other thermochemical cycles mainly due to its safety and high efficiency. UTC process includes three steps, in which only the first step is unique. Hydrogen production apparatus with hectogram reactants was designed in this study. The results showed that high yield hydrogen was obtained, which was determined by drainage method. The results also indicated that the chemical conversion rate of hydrogen production was in direct proportion to the mass of  $\text{Na}_2\text{CO}_3$ , while the solid product was  $\text{Na}_2\text{UO}_4$ , instead of  $\text{Na}_2\text{U}_2\text{O}_7$ . Nevertheless the thermochemical cycle used for hydrogen generation can be closed, and chemical compounds used in these processes can also be recycled. So the cycle with  $\text{Na}_2\text{UO}_4$  as its first reaction product has an advantage over the proposed UTC process, attributed to the fast reaction rate and high hydrogen yield in the first reaction step.

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## 1. Introduction

Increasing energy consumption, exhaustible nature of fossil fuels and their effects on environment have enhanced interest in production of hydrogen from thermochemical cycles. In this process, water is split into stoichiometric amounts of hydrogen and oxygen in a series of chemical reactions, with heat as the only energy source. Thermochemical cycles convert thermal energy into stored chemical energy (hydrogen and oxygen) directly, on which research largely began in the 1960s and 1970s and involved nuclear reactors [1,2] and solar collectors [3] as the energy sources. Numerous thermochemical cycles for splitting water have been proposed, and generally can be divided into two broad types: high-temperature two-step processes [4,5] (the temperatures are well above 1000 °C) and low temperature multistep processes [6,7], which is designed to operate below 1000 °C. Two-step processes involve simple reactions but the temperatures required are above 1000 °C. Low-temperature processes produce intermediates that can be complex, corrosive mixtures, eg. Iodine-sulfur (I-S) cycle [8].

Oak Ridge National Laboratory (ORNL) proposed a thermochemical cycle called UTC process in 2009 that involved non-

corrosive solids and also operated at a lower temperature 650 °C, with uranium [9] as its multi-valent moiety. The flow chart of this process and its full description was shown in literature [10], which allowed for the use of a broader spectrum of heat sources. Renewable energy has been considered to solve the energy crisis and sustainable development. But it is not sufficient to meet the global energy demand. Thus, attempts are being made to use nuclear energy as a supplementary energy source. Advanced nuclear power is clean and can provide a large-scale energy supply except for accidents. Thorium Molten Salt Reactor (TMSR), as one of reactor types in Generation IV reactors [11], has many advantages over uranium-based nuclear energy, such as their deterministic safety, sustainability, and nonproliferation. TMSR is a real peace application of nuclear energy, without resulting in nuclear proliferation. A design study of a 2 MW experimental Molten-Salt Reactor has been proceeding in our institute, and it has been developed to work with an outlet temperature of 700 °C, which is in line with the requirement of UTC process (Table 1). The objective of our work is to verify the UTC process, so as to make the best of outlet temperature of TMSR and abundant depleted uranium in china, which is considered as a by-product of the uranium enrichment process in the nuclear fuel cycle [12]. In this work, hydrogen was separated from other gases by selective membranes and pure hydrogen was obtained, while in previous work, reduction of CuO to copper metal, determined by weight loss, was the primary method used to measure the amount of reductant produced

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**Table 1**  
Comparison of hydrogen production methods using nuclear power.

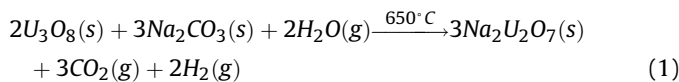
Hydrogen production	HTSE	Thermochemical cycles			
		I–S	Cu–Cl	Ca–Br	U–C
Energy input	Electricity + Heat	Heat	Electricity + Heat	Heat	Heat
Temperature (°C)	800	850	530	750	650
Key benefits	No pollution with renewable sources	Clean and sustainable			
Critical challenges	Low efficiency Capital costs Integration with renewable energy sources	Effective and durable materials of construction			

HTSE, High temperature steam electrolysis.

[9], which may include H<sub>2</sub> and CO.

## 2. Material and methods

U<sub>3</sub>O<sub>8</sub> used in this work was bought from China North Nuclear Fuel Co. Ltd (CNNFC), in which the uranium content and the percentage of major elements are shown in Table 2. All chemicals used were of analytical grade. The chemical equation of hydrogen production step was shown in Eq. (1). The reactants were prepared by mixing U<sub>3</sub>O<sub>8</sub> and Na<sub>2</sub>CO<sub>3</sub> with different molar ratios with IKA grinding machine.



Our new designed apparatus used in this work is shown in Fig. 1. It consisted of a steam generator, a hydrogen separation system, a high temperature rotary furnace and a glove box. Membrane separation technology had the advantages of low energy consumption, simple equipment, easy operation, no phase change during separation, good selectivity and suitable for laboratory-scale experiments, which was adopted in this study. H<sub>2</sub> yield was in direct proportional to the pressure of mixed gas and H<sub>2</sub> volume fraction in the mixed gas [13], so the hydrogen separation system consisted of a gas booster and a hydrogen purifier to increase the yield of hydrogen production. Palladium membrane was the key separation component in the hydrogen purifier, hydrogen could quickly pass through the membrane under pressure by means of dissolution-diffusion [14,15], while other impurity gases were trapped and discharged through the tail gas.

The reaction tube was made of Inconel 600 in case of corrosion. Vapor inlet part included a vapor inlet tube, an Ar inlet tube and a thermocouple placing tube, which monitored temperature in the tube. This part connected with the furnace tube by a rotating joint. In case of hydrogen leakage, a H<sub>2</sub> detector was installed over the equipment, the electricity and solenoid valve on the vapor inlet tube can be cut off once hydrogen was detected. The right side of furnace was linked with the glove box by a corrugated pipe, so it can tilt at any desirable angles within the range of 0–30 °C, which was convenient for injection and discharge of solid.

The experiment was performed on the equipment after each component operated regularly. Water (H<sub>2</sub>O) vapor was introduced

from the steam generator. The furnace was heated to the test temperature, with the typical ramp rate was 10 °C/min, the vapor stream was then charged into the tubular furnace and was maintained at the test temperature for several hours. Gases flew through a jacketed alkali solution tank and then a calcium oxide drying tower before they entered the hydrogen purification system. Gas pressure after the booster was maintained at 10 atm, then H<sub>2</sub> and effluent gases were collected in an aluminum foil gas sampling bag respectively. After the test was terminated, the equipment was swept by an Ar stream to collect remaining hydrogen. The solid product sample was collected in the glove box while the furnace tilted.

The composition of solid product was determined by X-ray diffraction methods. Qualitative analyses of all gas samples were performed on Agilent 7820 A gas chromatography, and then quantitative analysis of H<sub>2</sub> end gas was finished by drainage method. The chemical conversion rate was obtained by the analysis of the solid product, which was compared to the quantitative analysis result of H<sub>2</sub> end gas.

## 3. Results and discussion

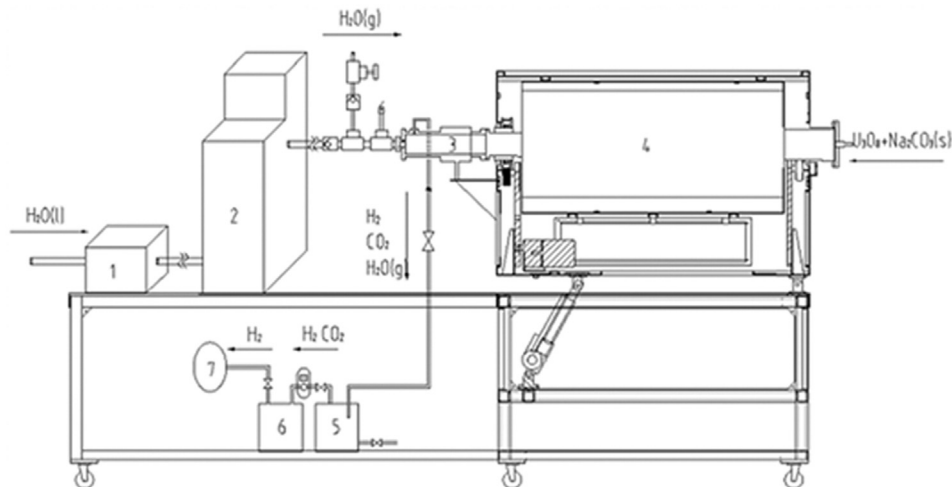
### 3.1. Gas-tightness test on the hydrogen product equipment

The reaction of hydrogen production proposed a challenge for the air-tightness of equipment, due to minimum molecular weight of hydrogen. An air-tight test was performed before experiments, which was performed by a vacuum pump. According to the final pressure in the equipment after 24hrs, gas leakage rate was calculated to be 6.5 Pa/h, under which leakage of H<sub>2</sub> could be ignored [16].

Hydrogen is inflammable and has explosive properties over wide ranges when mixed with air. Oxygen must be eliminated from the system also because U<sub>3</sub>O<sub>8</sub>, Na<sub>2</sub>CO<sub>3</sub>, and O<sub>2</sub> can react spontaneously to form Na<sub>2</sub>U<sub>2</sub>O<sub>7</sub> and reduce the H<sub>2</sub> yield. Inert Gas Ar was consumed for air displacement in the equipment after gas-tightness test. In general, the maximum allowable concentration of oxygen was not greater than 10% for most of inflammable substances [17], which must be controlled lower during operation. Vacuum deaeration was conducted three times until concentration of oxygen in the equipment decreased to 1\*10<sup>-6</sup>, which was calculated based on the methods presented in literature [18].

**Table 2**  
Uranium content and percentage of major elements in U<sub>3</sub>O<sub>8</sub>.

Sample NO.	U (%)	Concentration (μg/g U)								
		Cr	Fe	Ni	Si	Ca	Mg	Al	F	Cl
U <sub>3</sub> O <sub>8</sub> -317	84.81	<5	<10	<5	<10	<1	<5	<5	<10	<6
	H <sub>2</sub> O(%)	N	C	Zn	B	Cd	Sm	Eu	Gd	Dy
U <sub>3</sub> O <sub>8</sub> -317	0.06	<12	<22	<5	<0.1	<0.1	<0.02	<0.01	<0.02	<0.02



**Fig. 1.** The apparatus for hydrogen production step. 1. Constant-flow pump, 2. Steam generator, 3. Rotating joint, 4. Rotary furnace, 5. Condenser, 6. Hydrogen purification system, 7. Aluminium foil bag.

### 3.2. Effect of water flowrate on the temperature in the tube

Hydrogen production reaction was endothermic, so temperature in the tube had a tremendous effect on the process, which was studied in detail. Thermocouple monitored temperature in the tube in real-time, which extended to the center of thermostatic area. Because of inevitable heat loss during heat transfer, the temperature in the tube was lower than furnace set temperature. This temperature differences were studied at different furnace temperatures, which were kept at a certain temperature for more than 1 h. Results are shown in Table 3, which can be seen that there were a little differences between them, and this difference decreased as the furnace temperature increased.

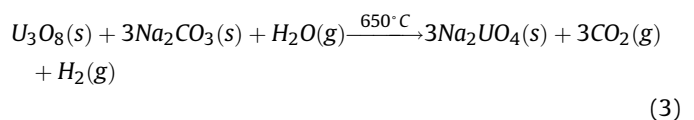
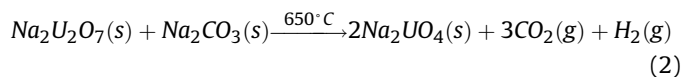
Steam was involved in this reaction, temperature of which was far below the furnace temperature. So it might have a fundamental effect on the temperature in the tube after its injection, which was also studied in this article. Steam was injected after the temperature in tube was steady. Results are shown in Table 4, with different water flowrate and furnace temperature. It indicated that temperature in the tube increased with the decrease of water flowrate at the same furnace temperature. However, the pressure of outlet steam was negative, which meant that steam was inclined to be condensed due to the long pipeline of inlet gas. And it might cause agglomeration of solid in the tube, which was bad for chemical conversion. So the flowrate of water must be large enough without increasing entrainment of solid. The actual temperature in the tube was about 650 °C, when water flowrate was 5 ml/min and furnace temperature was 720 °C, which were used as the optimum operation conditions later. These results also showed that the temperature differences between in and outside the tube increased significantly after the injection of steam, which were compared to the results shown in Table 3.

**Table 3**  
Temperature differences between in and outside tube.

Set temperature of furnace (°C)	Thermocouple detected temperature (°C)	$\Delta T$ (°C)
550	536.0	14.0
650	637.4	12.6
670	657.4	12.6
750	738.1	11.9
836	824.4	11.6

### 3.3. Effect of $U_3O_8$ and $Na_2CO_3$ molar ratio on the solid product

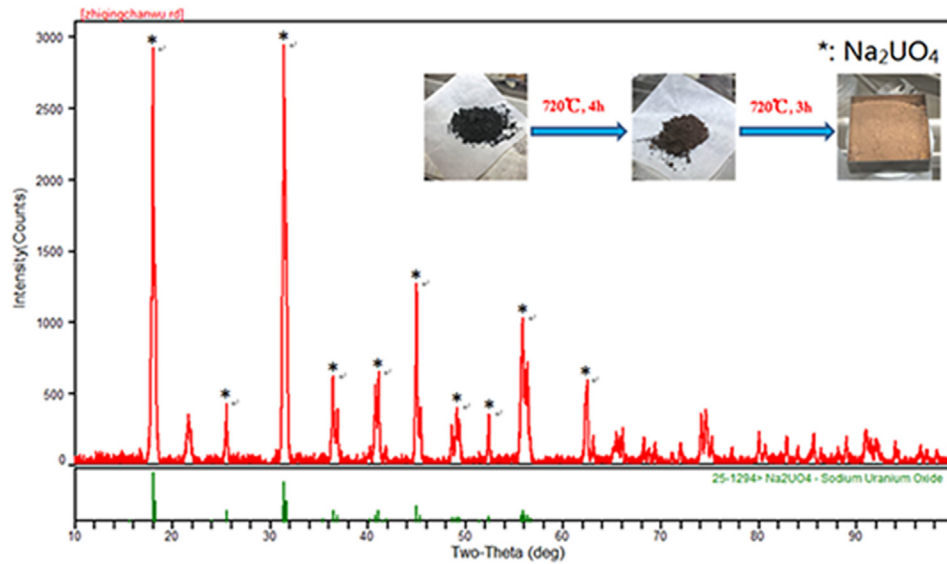
The reaction proceeded almost completely while  $Na_2CO_3$  was excessive, with molar ratio of  $U_3O_8$  and  $Na_2CO_3$  was 1:3.5. After reaction at 720 °C for 4 h, the solid product appeared in brown, finally orange product was obtained after another 3 h's reaction (Fig. 2). This product was analyzed by a Philips X'pert pro diffractometer with a 3 kW ceramic tube as the X-ray source (Cu KR) and an X'celerator detector, and its X-ray diffraction pattern is shown in Fig. 2. It can be seen that the solid product was  $Na_2UO_4$ , instead of  $Na_2U_2O_7$  proposed in the UTC process, and it was attributed to the following reaction of  $Na_2U_2O_7$  and excessive  $Na_2CO_3$  shown in Eq. (2), [19]. This result was consistent with the phase diagram of the system Na–U–O [20]. The chemical reaction was shown in Eq. (3), with  $Na_2UO_4$  as its product. However, despite different solid products,  $H_2$  theoretical output was identical. The orange solid product was quantitatively analyzed by TOC (Total Organic Carbon Analyzer), determining the remaining  $Na_2CO_3$ , while  $Na_2UO_4$  was measured by ICP-AES after it was resolved in  $(NH_4)_2CO_3$  solution, in which solubility of  $U_3O_8$  was little to be negligible. Based on the quantity of  $Na_2UO_4$  in the product, the conversion rate of hydrogen production reaction was about 93.6%.



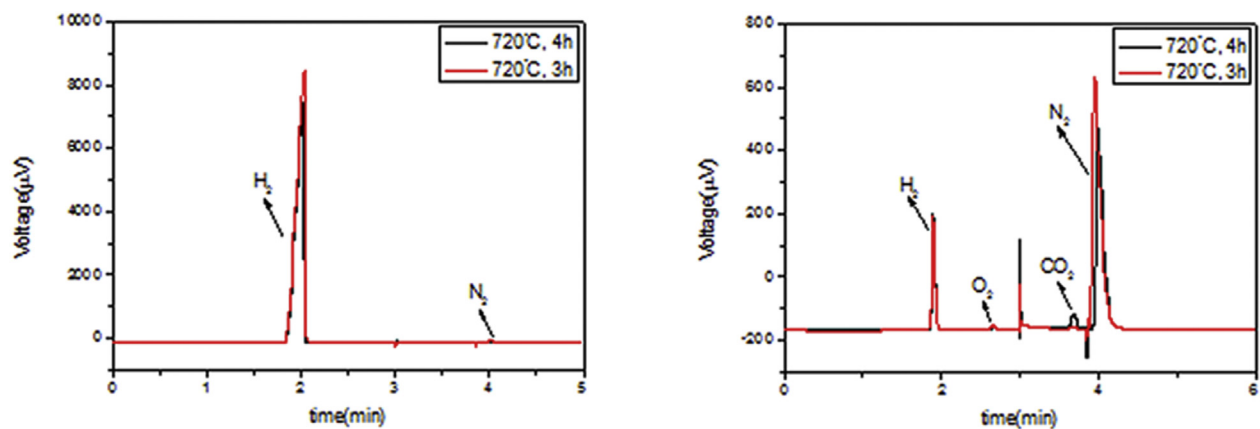
During two batches' experiments, three sampling bags (capacity of 1 L) of  $H_2$  end gas and two bags of effluent gas were collected, and their gas chromatograms are illustrated in Fig. 3, which can be seen that  $H_2$  purity was >99% and effluent gases were consisted of  $H_2O_2$ ,  $CO_2$  and  $N_2$ . The volume of  $H_2$  end gas was determined by drainage method after GC analysis. Theoretical volume of hydrogen produced based on the quantity of  $U_3O_8$  was calculated to be 2.4 L. Volume of hydrogen produced from the conversion rate of hydrogen based on produced  $Na_2UO_4$  was 2.25 L. While the volume of  $H_2$  end gas collected practically was 2.1 L, which was less than the volume of hydrogen produced based on produced  $Na_2UO_4$ . It means

**Table 4**  
Effects of water flowrate on temperature in the tube.

Flowrate of water (ml/min)	Temperature of outlet steam (°C)	Pressure of outlet steam (MPa)	Set temperature of furnace (°C)	Thermocouple detected temperature (°C)	$\Delta T$ (°C)
5	116.5	0.00	670	595.9	74.1
5	120.3	0.00	720	653.5	66.5
3	105.8	-0.04	720	669.1	50.9
5	119.1	0.00	750	692.4	57.6



**Fig. 2.** XRD pattern of the orange solid product. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)



**Fig. 3.** Gas chromatogram of H<sub>2</sub> end gas and effluent gas.

that H<sub>2</sub> wasn't collected completely, which was verified by H<sub>2</sub> peak in the gas chromatogram of effluent gas.

In order to obtain the desired Na<sub>2</sub>U<sub>2</sub>O<sub>7</sub> in the UTC process, molar ratio of U<sub>3</sub>O<sub>8</sub> and Na<sub>2</sub>CO<sub>3</sub> was changed to 1:1.8. After reaction at 720 °C for 21 h, the product was appeared in dark, which was analyzed by X-ray diffractometer. As illustrated in Fig. 4, the product was composed of U<sub>3</sub>O<sub>8</sub>, Na<sub>2</sub>UO<sub>4</sub> and Na<sub>2</sub>U<sub>2</sub>O<sub>7</sub> [21], so the reaction didn't proceed completely. It reacted in a more awkward way compared to the reaction with molar ratio of U<sub>3</sub>O<sub>8</sub> and Na<sub>2</sub>CO<sub>3</sub> was 1:3.5, which implied a lower H<sub>2</sub> yield because of lower H<sub>2</sub> volume fraction in the mixed gas. So quantity of Na<sub>2</sub>CO<sub>3</sub> had a fundamental effect on both the reaction rate of hydrogen

production and H<sub>2</sub> yield. If Na<sub>2</sub>U<sub>2</sub>O<sub>7</sub> is needed, according to the phase diagram of system Na–U–O, molar ratio of U<sub>3</sub>O<sub>8</sub> and Na<sub>2</sub>CO<sub>3</sub> should be further decreased, which means a much slower reaction rate. This study wasn't performed, while the effect of Na<sub>2</sub>UO<sub>4</sub> on the UTC process was investigated instead.

### 3.4. Thermodynamic analysis

In order to examine the spontaneity of reactions in Eq. (1) and Eq. (3), the changes of Gibbs free energy as a function of reaction temperature were calculated using the HSC 5.1 chemistry package [22], as plotted in Fig. 5. The thermodynamic approach predicted

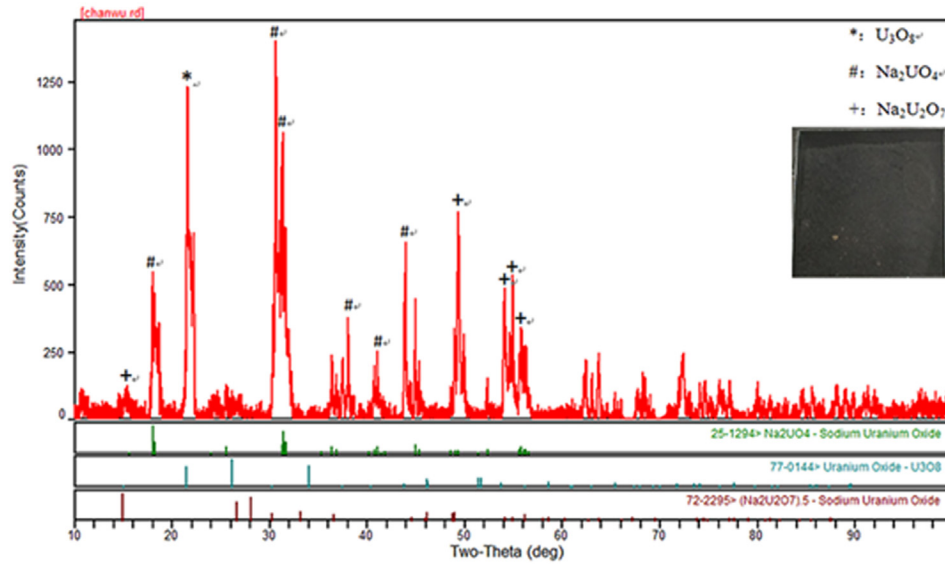


Fig. 4. XRD pattern of the solid product.

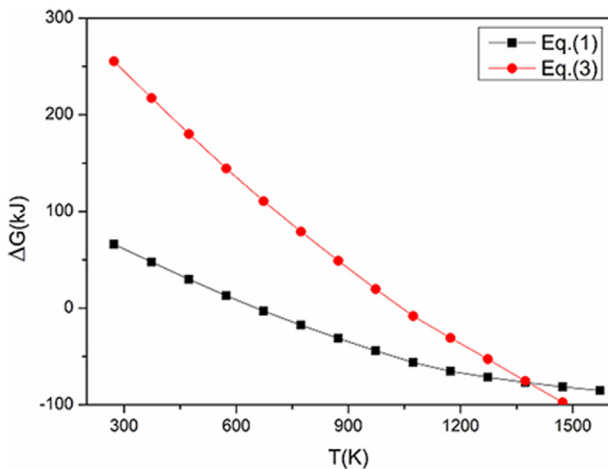


Fig. 5. Changes of Gibbs free energy of reactions as a function of temperature.

that both of these reactions were endothermic, and would occur much easier at high temperature. Reaction in Eq. (3) was difficult relative to Eq. (1), this difference became to disappear as temperature increased to 1373 K, after which reaction in Eq. (3) was much easier. In short, high temperature was beneficial to the reaction in Eq. (3). However, the process temperature must remain below the melting point of  $\text{Na}_2\text{CO}_3$  (1124 K). Hydrolysis of  $\text{U}_3\text{O}_8$  and  $\text{Na}_2\text{CO}_3$  leaved very little leeway for the process.

Thermodynamics also dictated operation at low pressure because of the positive variation in the number of moles of the gaseous species. However, the low pressure condition was impractical as well as inefficient for large scale production of hydrogen.

### 3.5. Verification of UTC process with $\text{Na}_2\text{UO}_4$ as the product of first step

According to the process proposed by ORNL, the second step must be realized and ammonium uranyl tricarbonate (AUC)

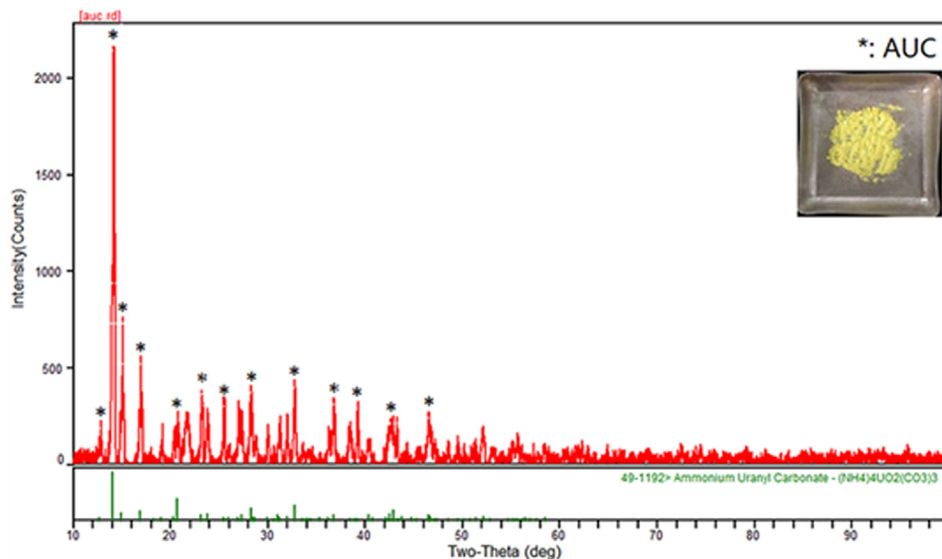


Fig. 6. XRD pattern of precipitate from Dissolved liquid of  $\text{Na}_2\text{UO}_4$ .

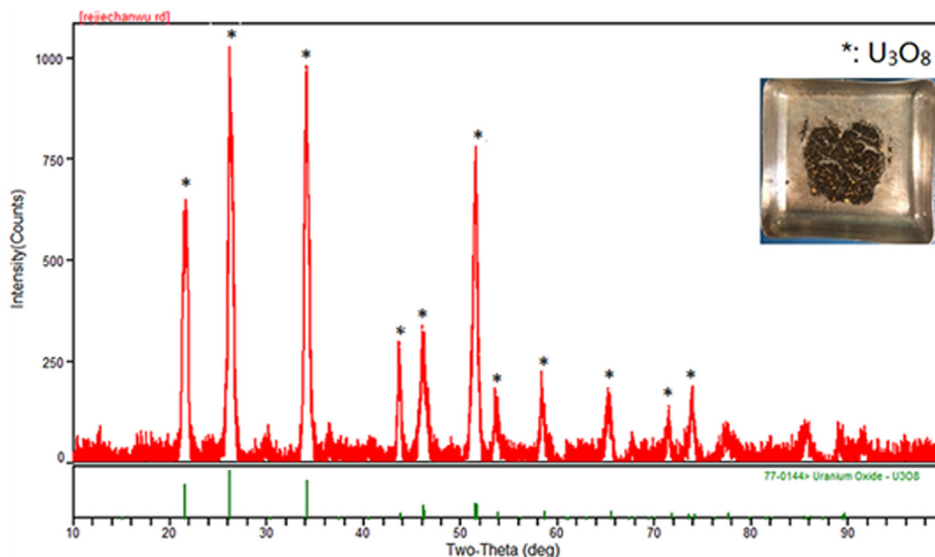


Fig. 7. XRD pattern of pyrolysis solid product.

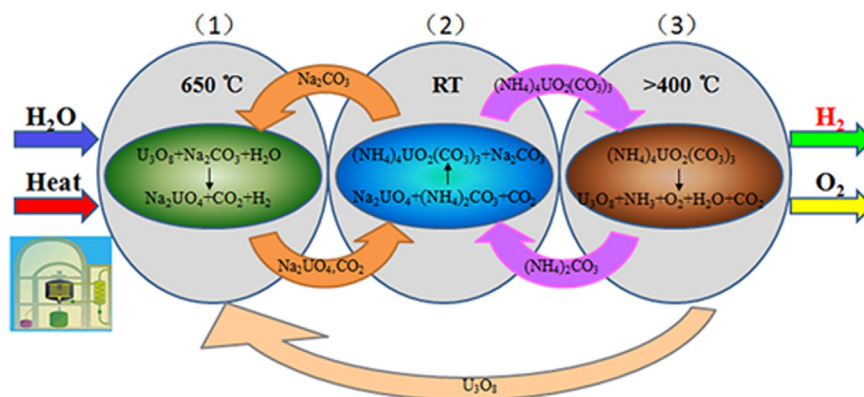


Fig. 8. Uranium thermochemical cycle with  $\text{Na}_2\text{UO}_4$  as product of the first step.

solution must be obtained as expected if the process needs to be cycled, so the solubility of  $\text{Na}_2\text{UO}_4$  in  $(\text{NH}_4)_2\text{CO}_3$  was investigated.

The reaction product was added into  $(\text{NH}_4)_2\text{CO}_3$  of certain molarity [23], in a round-bottom flask, with a temperature of  $50^\circ\text{C}$ ,  $\text{CO}_2$  flow was maintained during the experiment. Yellow clear solution was obtained after several hours' reaction, and then a certain amount of  $(\text{NH}_4)_2\text{CO}_3$  [24,25] was added to precipitate uranyl ion, which was performed at room temperature. The powder for this precipitation step was obtained after separating the solid by centrifugation and drying at  $60^\circ\text{C}$  in oven for 24 h. The powder was analyzed by X-ray diffractometer, with its XRD pattern illustrated in Fig. 6, it can be seen that the product was ammonium uranyl tricarbonate which verified the second reaction step in the UTC process. The yellow precipitate was decomposed at  $650^\circ\text{C}$  for 2 h, and it yielded black product, XRD pattern of which is shown in Fig. 7. It proved to be  $\text{U}_3\text{O}_8$ , a reactant in hydrogen production reaction, which was consistent with the third step in the UTC process.

Although the product of the first step, that was hydrogen production reaction, was  $\text{Na}_2\text{UO}_4$ , this cycle could be closed (Fig. 8), and substance in this process could also be cycled. However, hydrogen production rate was accelerated obviously, so did the efficiency of this cycle, and it also featured higher  $\text{H}_2$  yield. Therefore, the cycle with  $\text{Na}_2\text{UO}_4$  as its first reaction product had an

advantage over the former UTC process proposed by ORNL.

#### 4. Conclusions

The apparatus used for hydrogen production with hectogram reactants was designed, giving full consideration of the escape of solid powders and hydrogen. Pure hydrogen was obtained on this equipment and its output was about 2.1 L. The reaction rate of hydrogen production was in direct proportion to the mass of  $\text{Na}_2\text{CO}_3$ . However,  $\text{Na}_2\text{UO}_4$  was produced when excessive  $\text{Na}_2\text{CO}_3$  was added in this reaction, instead of  $\text{Na}_2\text{U}_2\text{O}_7$  as proposed in the UTC process. The following experiments revealed that  $\text{Na}_2\text{UO}_4$  can be resolved in  $(\text{NH}_4)_2\text{CO}_3$  and existed as AUC, which was converted into  $\text{U}_3\text{O}_8$  after pyrolysis reaction. Therefore, the new product  $\text{Na}_2\text{UO}_4$  can be used to propagate a continuous process in this closed cycle. The cycle with  $\text{Na}_2\text{UO}_4$  as its first reaction product has an advantage over the UTC process proposed by ORNL, attributed to the fast reaction rate and high hydrogen yield in the first reaction step.

#### Conflict of interest

None declared.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.net.2018.08.018>.

## References

- [1] J.E. Funk, R.M. Reinstrom, Energy requirements in the production of hydrogen from water, *Ind. Eng. Chem. Process Des. Dev.* 5 (3) (1966) 336–342.
- [2] J.E. Funk, Thermochemical production of hydrogen via multistage water splitting processes, *Int. J. Hydrogen Energy* 1 (1976) 33–43.
- [3] E.A. Fletcher, R.L. Moen, Hydrogen and oxygen from water, *Science* 197 (1977) 1050–1056.
- [4] S. Abanades, P. Charvin, F. Lemont, G. Flamant, Novel two-step SnO<sub>2</sub>/SnO water splitting cycle for solar thermochemical production of hydrogen, *Int. J. Hydrogen Energy* 33 (2008) 6021–6030.
- [5] P. Charvin, S. Abanades, G. Flamant, F. Lemort, Two-step water splitting thermochemical cycle based on iron oxide redox pair for solar hydrogen production, *Energy* 32 (2007) 1124–1133.
- [6] G. Beghi, A decade of research on thermochemical hydrogen at the Joint Research Center, Ispra, *Int. J. Hydrogen Energy* 11 (1986) 761–771.
- [7] L.C. Brown, Nuclear production of hydrogen using thermochemical water splitting cycles, in: *Proceedings of the International Congress on Advanced Nuclear Power Plants, Hollywood, Florida, June 13–19, 2002*.
- [8] G.H. Norman, L.C. Besencruch, L.C. Brown, Thermochemical Water-splitting Cycle, Bench-scale Investigations and Process Engineering, General Atomics Report, 1982. GA-A16713.
- [9] J.J. Ferrada, J.L. Collins, Carbonate thermochemical cycle for the production of hydrogen, in: *National Hydrogen Association (NHA) Annual Conference, Columbia, South Carolina, March 30–April 3, 2009*.
- [10] J.L. Collins, L.R. Dole, J.J. Ferrada, C.W. Forsberg, U.S. Patent 7,666,387 B2, U.S. Patent and Trademark Office, Washington, 2009.
- [11] J.E. Kelly, Generation IV International Forum: a decade of progress through international cooperation, *Prog. Nucl. Energy* 77 (2014) 240–246.
- [12] J.G. Arnason, C.N. Pellegrini, Depleted and enriched uranium exposure quantified in former factory workers and local residents of NL Industries, Colonie, NY USA, *Environ. Res.* 150 (2016) 629–638.
- [13] D.L. Xie, J.F. Yu, F. Wang, N. Zhang, Hydrogen permeability of Pd–Ag membrane modules with porous stainless steel substrates, *Int. J. Hydrogen Energy* 36 (1) (2011) 1014–1026.
- [14] T.L. Ward, T. Dao, Model of hydrogen permeation behavior in palladium membranes, *J. Membr. Sci.* 153 (1999) 211–231.
- [15] J. Han, I.S. Kim, K.S. Choi, High purity hydrogen generator for on-site hydrogen production, *Int. J. Hydrogen Energy* 27 (2002) 1043–1047.
- [16] G.F. Weston, *Ultrahigh Vacuum Practice*, 1985. Boston.
- [17] L. Tian, H.D. Qu, *Chemical Engineering Safety Technology*, 1984. Beijing, China.
- [18] W.M. Shen, Calculation of inert gas consumption for flammable gas displacement, *Oil Gas Storage Transp.* 15 (1) (1996) 20–22.
- [19] L.G. Stonhill, The thermal decomposition of the uranyl and sodium uranyl carbonates, *Anal. Chim. Acta* 23 (1960) 423–427.
- [20] R. Pankajavalli, V. Chandramouli, S. Anthonysamy, K. Ananthasivan, V. Ganesan, Thermochemical studies on the system Na–U–O, *J. Nucl. Mater.* 420 (2012) 437–444.
- [21] A.L. Smith, J.Y. Colle, P.E. Raison, Thermodynamic investigation of Na<sub>2</sub>U<sub>2</sub>O<sub>7</sub> using Knudsen effusion mass spectrometry and high temperature X-ray diffraction, *J. Chem. Thermodyn.* 90 (2015) 199–208.
- [22] HSC Chemistry V 5.1, Outokumpu Research, Finland, 2002.
- [23] A.L. El-Ansary, G.M.A. El-Wahab, E.E. Bayoumi, A.N. El-Said, Purification of Abu-Zenima wet crude yellow cake using alkaline leaching of U(VI), *Egypt. J. Petrol.* (2017). <https://doi.org/10.1016/j.ejpe.2017.08.003>.
- [24] K.M. Wu, Solubility of uranyl ammonium carbonate, *Atomic Energy Sci. Technol.* 3 (1961) 148–156.
- [25] S.J. Jiang, F.Y. Ren, *Nuclear Fuel Processing Engineering*, 1995. Beijing, China.