Evaluation of neutron activation of intermetallic matrices for dispersive nuclear fuel obtained by SH-synthesis

M.M. Balachov, O.Yu. Dolmatov, M.S. Kuznetsov, K.S. Kostyuchenko, N.O. Pimenov, A.A. Permikin

National Research Tomsk Polytechnic University, 30, Lenin Avenue, Tomsk Polytechnic University, Tomsk, Russia, 634050
E-mail: aap71@tpu.ru

Abstract. The paper describes a technique for obtaining intermetallic matrix materials based on Zr-Al and Ni-Al systems for dispersive nuclear fuel. This type of fuel is planned to be used in existing and future high-temperature nuclear reactors. Physical and mathematical modelling of the process of activation of matrices by neutron radiation showed that upon reaching the value of thermal neutron fluence of the order of $2.76 \cdot 10^{21}$ n·cm^{-2}, the activity of Zr-Al and Ni-Al matrices was $1.3 \cdot 10^{10}$ and $0.2 \cdot 10^{10}$ Bq/g, respectively. The analysis showed that in terms of exposure of irradiated fuel it is preferable to use a matrix based on the zirconium-aluminium system.

1. Introduction

Nowadays, uranium dioxide (UO_2) is used as nuclear fuel in the majority of nuclear reactors. The main disadvantage of this nuclear fuel type is the low thermal conductivity coefficient which is equal to 2.8 W/(m·K) at operating temperatures [1]. Specifically, temperature gradients take place in fuel compositions which have negative influence on strength characteristics of fuel pellets and set a limit for the temperature operation mode of nuclear facility. So, the NPP work efficiency upgrading problem is connected with a development of new improved nuclear fuel types meeting the strict safety requirements. Nowadays, one of the most advanced nuclear fuel type is dispersion nuclear fuel (DNF).

DNF consists of two phases. The first phase is a non-active matrix; the second one contains fuel particles dispersed in the matrix. Schematically the dispersion nuclear fuel microstructure is presented in figure 1. The correct choice of micro particles size and fraction allows localizing all radiation damages in the non-active matrix, so that DNF high radiation hardness is ensured.

The compounds with high thermal conductivity coefficient, such as metals, intermetallides, graphite, can be used as matrix in dispersion nuclear fuel what ensures high heat removal from the fuel composition in comparison with traditional fuel types. Ni and Zr are in a row of transition metals; alloys based on them have unique thermal and physical properties what offers wide opportunities for using them as a matrix for dispersion nuclear fuel [2, 3]. In order to produce such fuel type standard metallurgy methods are used, such as smelting, casting, blanking and piercing, etc. [4]. However these methods have a number of disadvantages, such as expensive production and difficult technological performance. The method of self-propagating high-temperature synthesis (SHS) does not have these disadvantages. The SHS is an advanced material engineering technology based on the ability of some chemical elements and compounds to undergo exothermic reactions [5].

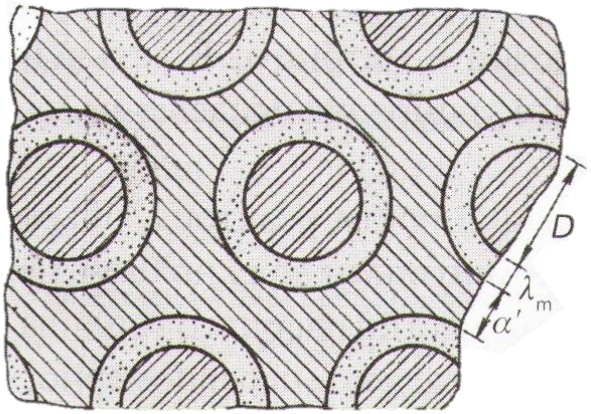

Figure 1. Dispersion nuclear fuel composition structure scheme: D – fuel particle diameter; λ_m – damaged matrix material thickness; α' – distance between damaged matrix layers.

The SH-synthesis consists of following stages:

- heat is added pointwise to a pressed product consisting of reagents mixture;
- a reagents thin layer is heated and undergoes an exothermic reaction;
- the next sample layer is heated to synthesis temperature imitation due to thermal conductivity, after that new exothermic reactions series starts;
- so, a reaction wave propagates over whole sample.

2. Experiment

The SH-synthesis of intermetallic matrix consists of several stages. In the early stage an initial reagents powder mixing is carried out in the cubic mixer AR 403 All-Purpose Equipment. Then a batch pressing is carried out using the hydraulic press P338. The sample preparation parameters are presented in table 1.

Table 1. DNF matrix preparation parameters.

Parameter	System of Zr-Al	System of Ni-Al
Powder grades	PAP-2, ZRCP-1	PAP-2, CAS 7440
Mixing time, min	30	30
Extrusion pressure, MPa	30	30
Pressing duration, min	15	15
Billet diameter, mm	30	30
Billet height, mm	10-20	10-20

Batches were prepared on the basis of following exothermic reactions:

$$\begin{cases} \mathrm{Ni + Al \rightarrow NiAl} \\ \mathrm{Ni + Al + NiAl \xrightarrow{\downarrow T} 2NiAl} \end{cases}; \qquad (1)$$
$$\mathrm{3Zr + Al \rightarrow Zr_3Al.}$$

The SH-synthesis of material was initiated after pressing by the local heat supply. The synthesis was carried out in a special reactor in technical vacuum. The synthesis was carried out in technical vacuum on the test bed for pyrometric research of SH-synthesis process including a SHS-reactor. The SHS-reactor is presented in figure 2. The synthesis process is pictured in figure 3.

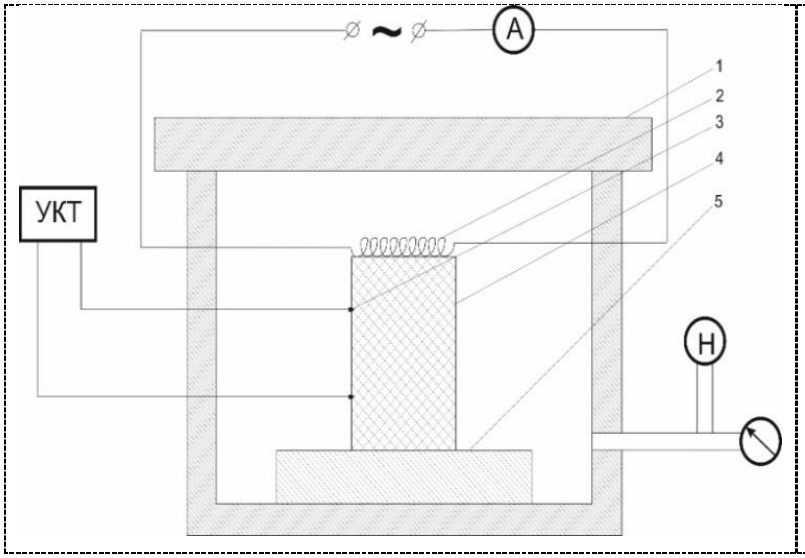

Figure 2. Experimental facility for SHS-materials obtaining: 1 – reactor head; 2 – tungsten filament; 3 – thermocouple complex; 4 – synthesized sample; 5 – nonflammable platform.

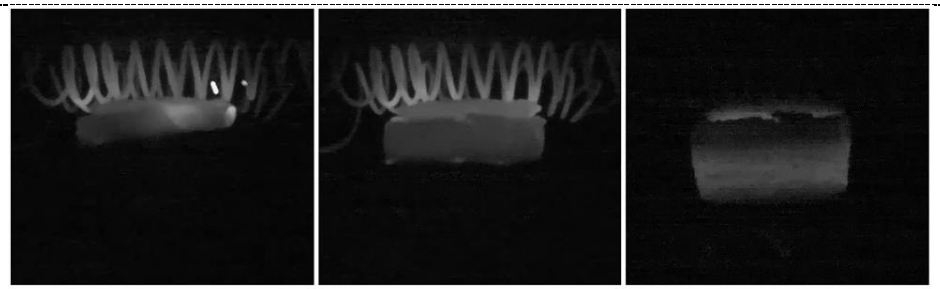

Figure 3. Self-propagating high-temperature synthesis.

3. Results and Discussion

The synthesis process was controlled by tungsten-rhenium thermocouple complex. The temperature regime of reaction course is presented in figure 4.

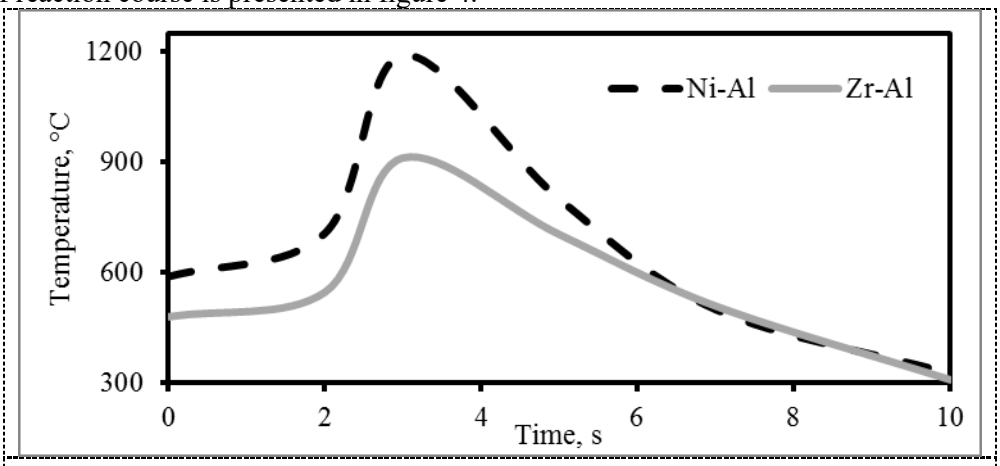

Figure 4. Synthesis temperature regime.

The maximum matrix synthesis temperature for a system of Ni-Al is equal to 1200 °C. A system of Zr-Al has lower thermal effect with the maximum temperature of 900 °C.

After the synthesis samples were X-ray phase analyzed in the diffractometer Shimadzu XRD-7000. The phase composition analysis was carried out using the data base PDF 4+, as well as the full-profile

analysis program POWDERCELL 2.5. The results of X-ray phase analysis are presented in figures 5 and 6.

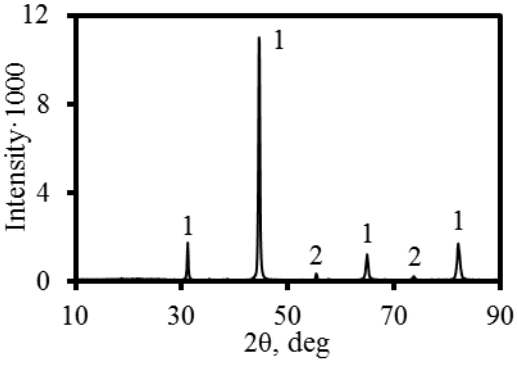

Figure 5. Phase composition of sample based on the system of Ni-Al: 1: NiAl – 99%; 2: Al_2O_3 – 1%.

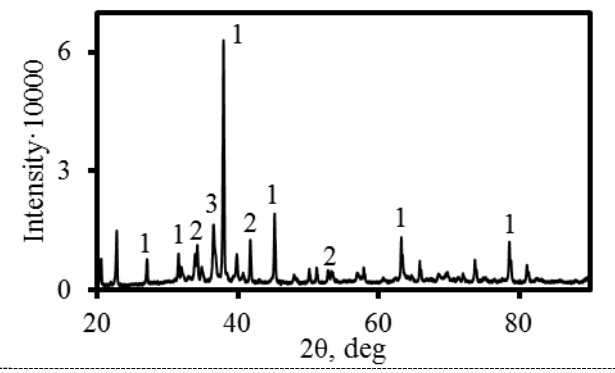

Figure 6. Phase composition of sample based on the system of Zr-Al: 1: Zr_3Al – 74%; 2: $ZrAl_2$ – 19%; 3: αZr – 7%.

It will be seen that the nickel aluminide sample contains 99 % of target phase. The zirconium aluminide sample contains 7 % of unreacted alpha-zirconium what stipulates the necessity of further SH-synthesis controlling research in order to make experimental conditions for decreasing of α-Zr content and increasing of Zr_3Al content.

The important problem is a physical and mathematical modeling of DNF matrix activation process by neutron radiation while in fuel operation in a nuclear reactor. Therefore, for the purpose of this work the model of matrix nuclide inventory changing based on the systems of Zr-Al and Ni-Al obtained by the SH-synthesis method was developed.

Neutrons capture reactions and nuclei beta-decay reactions were considered in this model because they are the most important in terms of radioactive isotopes generation.

The mean of thermal neutrons flux in the reactor WWER-1000 at nominal power is $2.66 \cdot 10^{13}$ n·cm^{-2}·s^{-1}. The radiant exposure of thermal neutrons for a four year period is $2.76 \cdot 10^{21}$ n·cm^{-2} [6]. The matrix activation calculation is focused on this radiant exposure value.

Nuclear reactions arising in the matrix take the following form:

$$X_Z^A + n_0^1 \xrightarrow[(n,\gamma)]{\sigma_\gamma} X_Z^{A+1};$$
$$X_Z^A \xrightarrow[\beta-]{T_{1/2}} X_{Z+1}^A. \quad (2)$$

In this case every isotope concentration changing is described by the expression:

$$\frac{dN(t)_{A,Z}}{dt} = -N(t)_{A,Z} \cdot \sigma_{A,Z} \cdot \Phi - N(t)_{A,Z} \cdot \lambda_{A,Z} + N(t)_{A-1,Z} \cdot \sigma_{A-1,Z} \cdot \Phi + N(t)_{A,Z-1} \cdot \lambda_{A,Z-1}, \quad (3)$$

где N – nuclei number; t – time; σ – radiation capture cross section; λ – decay constant; Φ – neutron flux.

Wolfram Mathematica was used for the calculation. It is worthwhile noting that Al^{28} and Mn^{56}, Cu^{64} make a huge contribution to a matrix radioactivity accumulation because of short half-life. However the radioactivity of these isotopes is leveled during a few score of hours while radiation is detected. So that given nuclei are not considered in this model. The graph of matrix radioactivity dependence from a neutron radiant exposure is presented in figure 7.

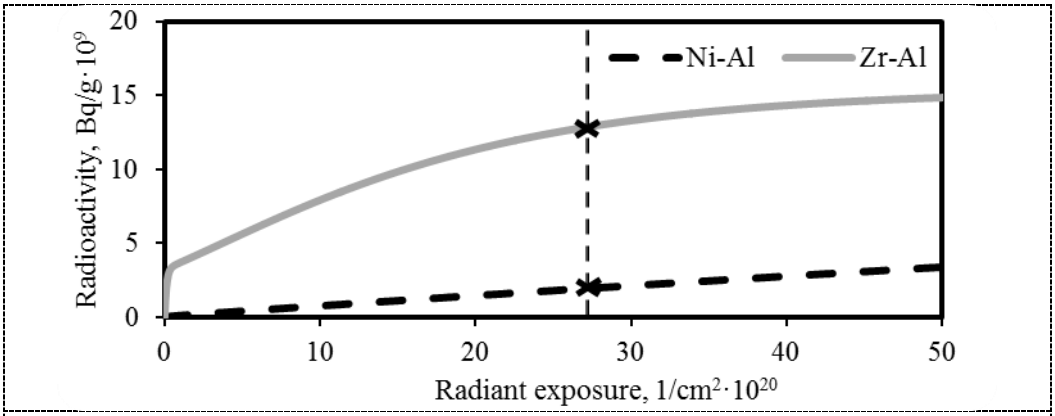

Figure 7. Sample radioactivity accumulation.

Analyzing the obtained dependences it was concluded that the radioactivity of Zr-Al and Ni-Al matrix is equal to $1.3 \cdot 10^{10}$ Bq/g and $0.2 \cdot 10^{10}$ Bq/g when the neutron radiant exposure in a fuel assembly of WWER-1000 reactor reaches the maximum value. When the neutron radiant exposure reaches the value of $5 \cdot 10^{21}$ n·cm^{-2} the radioactivity of Zr-Al matrix reaches the stationary level of $1.5 \cdot 10^{10}$ Bq/g. However, the radioactivity of Ni-Al system reaches the stationary level at the much higher radiant exposure of ~ 10^{22} n·cm^{-2}. The use of nickel-aluminum matrix is more reasonable in terms of operation lifetime.

The next work stage was to determine a decrease dynamics of matrix radioactivity after a radiation detecting. The results are presented in figure 8.

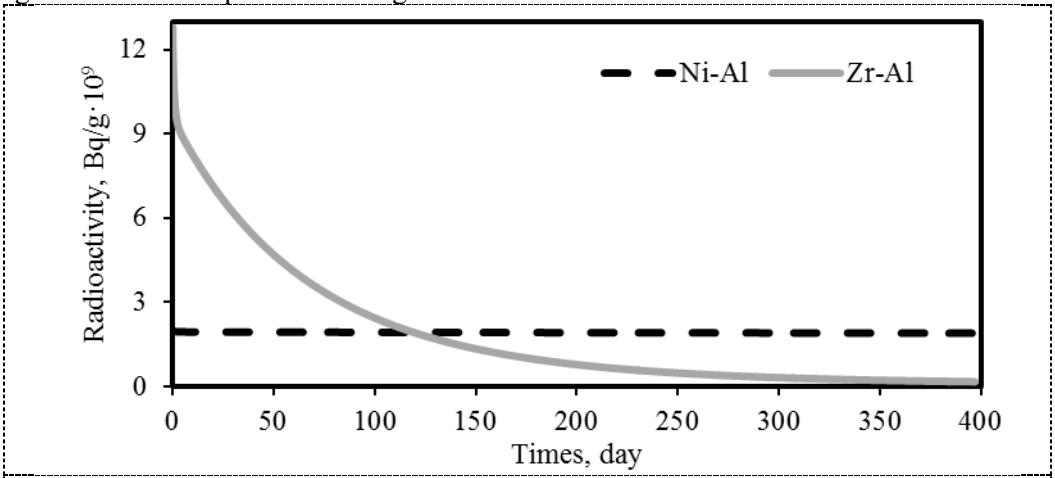

Figure 8. Decrease dynamics of matrix radioactivity.

It can be observed that Ni-Al matrix radioactivity decreases slightly during one year. This fact is stipulated for a high quantity accumulation of Ni^{63} isotope having the half-life of 100.1 years in the sample.

Analyzing the represented data it can be concluded that the use of Zr-Al matrix is more reasonable in terms of radioactive waste storage, reprocessing and utilization. It is related the fact that the radioactivity decrease intensity of Zr-Al matrix system is well above than the equivalent quantity of Ni-Al matrix system while radiation is detected.

4. Conclusion

This paper presents the production process of DNF intermetallic matrix based on Zr-Al and Ni-Al systems using a self-propagating high-temperature synthesis method. The use of this method allowed reaching the target phase mass content of 90 % in the samples.

The model which allows defining the changing of DNF matrix nuclei inventory while using in thermal nuclear reactors was developed. The analysis has shown that the use of matrix materials on the basis of Zr-Al system is more reasonable in terms of spent nuclear fuel radioactivity.

This work allows forecasting of matrix residual radioactivity after its removal from a radiation zone. It is necessary for further radiation hardness research of intermetallic matrix obtained by using the SHS method.

References

[1] Kotelnikov R B, Bashlykov S N, Kashtanov A I and Menshikov T S 1978 *High Temperature Nuclear Fuel* (Moscow: Atomizdat) p 432
[2] Kolobov Yu R, Kablov E N, Kozlov E V, Koneva N A, Povarova K B, Grabovetskaya G P, Buntushkin V P, Bazyleva O A, Muboyajjyan S A and Budinovsky S A 2008 *Structure and Properties of Intermetallic Materials with Nanophase Hardening* (Moscow: National University of Science and Technology MISiS) p 328
[3] Kablov E N, Ospennikova O G and Bazyleva O A 2010 *J. Engine* **70** 22-4
[4] Alekseev S V, Zaitsev V A and Tolstoukhov S S 2015 *Dispersion Nuclear Fuel* (Moscow: Technosphere) p 248
[5] Merzhanov A G 2006 *J. Russian J. of Non-Ferrous Metals* **5** 5-8
[6] Belozerov V I, Zhuk M M, Kuzina Yu A and Ternovykh M Yu 2014 *Physics and Operating Conditions of the WWER-1000 Reactor* (Moscow: MEPhI) p 288