Manufacturing and STA-investigation of witness-samples for the temperature monitoring of structural steels under irradiation

O N Sevryukov, V T Fedotov, A A Polyansky, S A Pokrovski and R S Kuzmin
National Research Nuclear University MEPhI (Moscow Engineering Physics Institute)
31 Kashirskoe highway, 115409 Moscow, Russia
E-mail: sevr54@mail.ru

Abstract. The object of investigations was alloys based on lead and cadmium used as fuse monitors to control the maximum irradiation temperature (fuse temperature monitors, FTM) of samples from structural steels under irradiation in a research reactor IR-8. The result of the work was selected and tested initial materials for production of alloys. A technological scheme of the production of alloys for FTM has been developed and experimental studies of the properties of these alloys have been carried out.

1. Introduction
To develop the technologies of increasing the service life of heat resistant steels, operated for a long time under extreme conditions as a part of critical constructions on the basis of an improved complex technique of certification of the service properties conditioned by the formation and transformation of the nanostructure during irradiation, it was necessary to develop compositions and the manufacturing technology of alloys for FTM in the form of rods with a diameter of about 3 mm and a melting temperature changing in the 298–314 °C range[1,2]. A technological scheme providing the production of rods with a specified chemical composition was proposed and the technique for measuring melting temperatures, which allows certifying FTM-alloys with a required accuracy, was improved.

2. Development of a method to produce alloys with a specified composition
The technological scheme of producing alloys is shown in figure 1. A vertical furnace with resistive heating was used as the basic unit of the equipment. The protection of the melt from oxidation is provided by an argon atmosphere and the melting temperature is controlled by a thermocouple that is connected with a power regulator (not shown in the scheme).

The filling rate of the quartz feeder is regulated by a rotameter, which allows choosing the optimum speed of solidification of the melt. The operability of the technological scheme at the stage of development was confirmed by obtaining samples of rods from a lead-indium alloy with a specified composition Pb–(8–10 wt. %)In
The technological scheme for production of alloys with a melting temperature of 250–350 °C.

Figure 1. The technological scheme for production of alloys with a melting temperature of 250–350 °C.

The geometric and thermo physical characteristics of the alloy obtained correspond to specified requirements. Lead of analytical reagent grade (ARG), cadmium (ARG), indium of the In0 grade, and tin of very high purity (VHP) were used as initial materials to produce a pilot batch of FTM-alloys [3]. To avoid contamination of the alloys by lead oxides, the melt of lead was preliminarily purified in an argon atmosphere. Alloys of six compositions: No.1 - Cd-(5–10)In; No.2 - Pb-(5–8.5)In; No.3 - Pb-(8–10)In; No.4 - Pb-(2–5)In; No.5 - Pb-(0.5–1.5)Sn and No.6 - Pb-(1–2)In, were made.

3. Measuring the melting temperatures and determining the chemical composition of experimental alloys.

To determine the melting temperature, an installation of the synchronous thermal analysis STA 409 CD of the Firm “Netzsch” (Germany) was used. To measure melting temperatures, a furnace with a Pt-Rh heater and a sample holder TG / DSK with a thermocouple of the type S (Pt–Pt-10%Rh) were used. The heating rate was 10 K/min. Tests were conducted in a dynamic helium atmosphere at a flow rate of 70 ml/min. Samples were placed in an Al₂O₃ crucible with "pierced" lids, and an empty crucible with a lid was used as the "standard".

To determine the influence of various factors (the contact with the crucible and the mass of samples) on the results of the experiment, preliminary measurements on samples of the alloy Pb-5%In were carried out. Three discs of various thicknesses were cut from the initial cylindrical rod of the alloy. Masses of the samples were 21.78, 52.18 and 101.50 mg. The melting of each sample was performed three times.

The results obtained show that the melting temperature shifts towards the area of lower values with increasing the sample mass. Additionally, for samples with large masses after the first melting, there is apparently a change of the contact with the crucible and the temperature of the extrapolated start for repeated meltings shifts towards lower values. Thus, to increase the measurement accuracy, it is necessary to preliminarily prepare (polish) the sample surface to ensure a good contact with the bottom of the crucible. It is also necessary to use samples with similar masses and perform a temperature calibration of the installation using standards with similar geometry and mass.

Determination of the melting temperatures was performed using alloys of six compositions. To preliminarily estimate the stability of the compositions obtained, several rods of each alloy were randomly selected for each composition. Further, samples for tests were produced from an arbitrary place of the rod.

For example, the alloy No. 6 shows a good reproducibility of the melting temperatures of samples of the same composition. Thermograms of the tests of five samples are presented in figure 2.
Figure 2. Thermograms of samples of the alloy No. 6 with masses of: 1, 44.71 mg; 2, 63.50 mg; 3, 63.79 mg; 4, 76.60 mg; 5, 78.22 mg.

Based on these results, the stability of the rest of alloys was evaluated by three samples. The test results of alloys No. 2, 3, 4 and 5 are shown in figures 3-6, respectively.

Figure 3. Thermograms of samples of the alloy No. 2 with masses of: 1, 36.78 mg; 2, 40.51 mg; 3, 30.80 mg.

Figure 4. Thermograms of samples of the alloy No. 3 with masses of: 1, 53.85 mg; 2, 58.11 mg; 3, 65.58 mg.

Figure 5. Thermograms of samples of the alloy No. 4 with masses of: 1, 34.32 mg; 2, 41.75 mg; 3, 58.28 mg.
Figure 6. Thermograms of samples of the alloy No. 5 with masses of: 1, 42.33 mg; 2, 45.79 mg; 3, 66.39 mg.

These results testify the stability of the composition and the manufacturing technology of alloys. The average difference of the melting temperatures for various samples of alloys of the same composition does not exceed 0.3 K, except for the alloy No. 2, where it reaches 0.7 K.

Special attention is given to the alloy No. 1. The results of its differential thermal analysis are presented in figure 7.

Figure 7. Thermograms of samples of the alloy No. 1 with masses of: 1, 37.81 mg; 2, 28.97 mg; 3, 13.84 mg.

As shown in the thermograms (figure 7), the alloy No. 1 undergoes several transformations at temperatures of 125, 160 and 187 °C that terminate at the temperature of 290 °C at which, presumably, the whole sample transfers into the liquid phase.

In this case it is difficult to accurately determine the complete transformation temperature because it will be significantly influenced by the heating rate and the sample mass. Considering the above said and the fact that the liquid phase appears in the alloy already at the temperatures of ~ 130 °C, the choice of the alloy of the given composition as a temperature indicator is questionable.

To ensure the best measurement accuracy, the thermocouple of the sample holder was calibrated under specified conditions using the melting temperatures of standard substances such as In, Bi and Zn. The calibration results are presented in Table 1.

Table 1. The results of determining the melting temperatures of standard materials.

| Material | Mass (mg) | Measured value of the melting temperature (°C) | Standard value of the melting temperature (°C) |
|----------|-----------|-----------------------------------------------|-----------------------------------------------|
| In       | 23.44     | 157.1                                         | 156.6                                         |
| Bi       | 54.64     | 272.0                                         | 271.4                                         |
| Zn       | 15.26     | 420.8                                         | 419.5                                         |
It follows from Table 1 that the melting temperatures measured for standard substances are slightly overrated and, as follows from figure 9, the dependence between the measured and standard values is linear.

![Figure 9](image.png)

**Figure 9.** The dependence between the measured and standardized melting temperatures of standard substances.

Using the calibration obtained, repeated measurements of the melting temperature of one sample for each alloy were made. The resulting melting temperatures of the alloys are shown in table 2.

**Table 2.** The melting temperatures of alloys.

| No. | $T_{\text{melting}}, ^{\circ}C$ |
|-----|-------------------------------|
| Alloy No. 1 | 289 ± 2 |
| Alloy No. 2 | 297 ± 1 |
| Alloy No. 3 | 299 ± 1 |
| Alloy No. 4 | 302 ± 1 |
| Alloy No. 5 | 309 ± 1 |
| Alloy No. 6 | 311 ± 1 |

4. Conclusion

Compositions have been selected in the systems of lead-indium, cadmium-indium and lead-tin and a technological scheme for the production of rods with a diameter of 3 ± 0.1 mm has been proposed. The melting points of alloys have been measured by a synchronous thermal analysis. Based on a complex of the investigations performed, the compositions of alloys have been clarified and alloys in the form of rods, providing the temperature control in the temperature range of 289–311 °C with a precision of ± 1–2 °C, have been made.

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References

[1] Rabu B, Pagano C, Boucenna A, Addes et al 1999 *Journal of Aerosol Science* **30** (1) 109

[2] Xiao H, Long C, Chen L and Liang B 2013 *Proc. 21st International Conference on Nuclear Engineering, ICONE 2013 (Chengdu, China)* I article number V001T02A027

[3] Dubourg R, Austregesilo H, Bals C et al 2010 *Progress in Nuclear Energy* **52** (1) 108