**ON MEASUREMENT OF OXYGEN CONCENTRATION**

**IN SODIUM WITH PLUG INDICATOR**

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**Abstract**

## The paper discusses the possibility of oxygen concentration measurement in sodium. The “plugging temperature” is normally measured by means of a plug indicator, and it is supposed to be equal to the temperature of sodium saturation with impurities. Then the concentration of oxygen is determined using an equation of oxygen solubility in sodium. As a rule, Noden´s equation is used. However, solubility of oxygen in sodium depends on the presence of other impurities in it. Therefore, there are many equations of oxygen solubility in sodium proposed by different authors under different conditions. Currently, at least 21 equations are known. The difference of these equations reaches ten times and more at a plugging temperature of 250-110°C. The authors of this paper have compared these equations and found out that almost all the equations converge at a temperature of 558 K and give a logarithm of concentration 1.957. Since this point satisfies all the equations, it can be used for calibration of plug indicators, and the equation of oxygen solubility in sodium can be obtained that is adequate to a real facility and operating conditions. The equation allows the oxygen concentration to be correctly determined.

## INTRODUCTION

## Plug indicators of impurities in liquid metal coolants are widely known and used at many experimental facilities, as well as in sodium-cooled nuclear reactors [1, 2]. The plug indicator basically consists of a flow meter, a cooler, a thermocouple and a throttle with a small flow area. The plug indicator operates as follows. The liquid metal temperature at the throttle inlet is lowered by means of a cooler, and the change of the flow rate and temperature is registered. When the temperature falls to saturation temperature, an impurity crystallization begins, and the flow area of the throttle decreases. As a result, the liquid metal flow rate falls as well. At the beginning of the flow rate reduction, the liquid metal temperature is registered as the ‘plugging temperature’, and it is considered as saturation temperature. Then, the saturation concentration is determined using the known solubility equation for the particular impurity (for example, oxygen) in the particular liquid metal (for example, sodium).

Figure 1 illustrates the results of one of the plugging temperature experiments at the IPPE experimental sodium facility.

*FIG. 1. Variation of sodium temperature and flow rate in the course of plugging temperature determination*

The beginning of the flow rate decrease is shown by a vertical line in the figure. The sodium temperature at the throttle inlet at that time was 152°C. It is considered as saturation temperature. Then the cooler was turned off and the indicator heater was turned on. When the sodium temperature reached 152°C again (dashed line), an increase of the flow rate, i.e. impurity dissolution, began. As Figure 1 shows, the plug indicator operates reliably at very low flow rates (as low as 21 *l/h*), therefore, it is small in size, and its power consumption for sodium pumping is low, and the power required for cooling is also low.

1. SOLUBILITY EQUATIONS ANALYSIS

The general view of the equation of oxygen solubility in sodium, i.e. the correlation between saturation concentration and temperature, is as follows:

|  |  |
| --- | --- |
| , | (1) |

where *C* is the saturation concentration, *T* – temperature, *A, B* – constant coefficients.

Many specific equations are known now. They were obtained by different authors and differ in the coefficients *A*, and *B*. These coefficients for the 21 best known equations taken from [3-9, 12] are presented in Table 1.

TABLE 1. EQUATIONS FOR SOLUBILITY OF OXYGEN IN SODIUM

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| № | *А* | *В* | Temperature, К | Year, source |
| 1 | 5.710 | 2043 | 425 – 822 | 1950 [3] |
| 2 | 4.080 | 1120 | 383 – 828 | 1951 [3] |
| 3 | 5.249 | 1176 | 400 – 813 | 1954 [3] |
| 4 | 5.054 | 1691 | 429 – 682 | 1955 [3] |
| 5 | 5.266 | 1816 | 388 – 778 | 1956 [3] |
| 6 | 5.129 | 1791 | 387 – 565 | 1960 [3] |
| 7 | 6.394 | 2518 | 428 – 613 | 1965 [3] |
| 8 | 5.210 | 1777 | – | 1965 [3] |
| 9 | 4.937 | 1652 | 403 – 753 | 1966 [3] |
| 10 | 8.250 | 3499 | – | 1966 [5] |
| 11 | 6.1217 | 2359 | 425 – 573 | 1968 [3] |
| 12 | 6.550 | 2600 | – | 1968 [3] |
| 13 | 6.239 | 2447 | – | 1968 [6] |
| 14 | 5.140 | 1910 | – | 1969 [4] |
| 15 | 7.076 | 2865 | 583 – 723 | 1971 [3] |
| 16 | 6.318 | 2355 | 373 – 673 | 1972 [3] |
| 17 | 6.131 | 2362 | – | 1972 [3] |
| 18 | 6.16±0.067 | 2387±35,3 | – | 1972 [7] |
| 19 | 6.2571 | 2444.5 | 383 – 823 | 1973 [8] |
| 20 | 6.02 | 2320 | – | 1982 [9] |
| 21 | 5.52 | 1900 | 433 – 598 | 1997 [12] |

Within the plugging temperature range of 250÷110 °C, which is relevant for the existing and future sodium-cooled reactor facilities, these equations give the values of concentration that differ by an order of magnitude. This difference primarily results from the presence of other impurities in sodium, such as hydrogen and elements of structural materials (Fe, Cr, Si, and others). They interact with sodium and oxygen and affect the solubility of oxygen [3]. The authors of the equations, as a rule, do not give information on the content of impurity elements and other conditions under which the equations were obtained. Sometimes the temperature interval of their experiments is unknown.

In addition, the nomenclature of impurities in sodium changes in the course of the facility operation. Therefore, at present, there is no (and, apparently, cannot be) a universal, "true" equation of oxygen solubility in sodium that is valid in each particular case.

The data of Table 1 are presented graphically in Figure 2 as a logarithm of mass concentration (ppm) versus inverse temperature (K). In the interval of 2.2 < 103/*T* < 2.6 they differ by an order of magnitude. That is why the plug indicator cannot be a measuring device to measure concentration. Even if calibrated at an experimental facility with a known error, it may produce unreliable concentration measurement results when it is transferred to another facility.

However, as Figure 2 shows, the graphs of all the equations, with the exception of two, have a minimum divergence in a narrow region of 1.7 ˂ 103/*Т* ˂ 1.8. In this range the average value of concentration logarithm over 19 equations for each temperature has a root mean square (RMS) uncertainty not more than ± 4%. Equations 3 and 14 deviate from the average value by more than three RMS and may be excluded as invalid. Therefore, the region with the coordinates 1.7 ˂ 103/Т ˂ 1.8, where 1.7 < lg*С* < 2.14, is valid for all the particular cases presented in Figure 2 with a high degree of probability (19 out of 21).

*FIG. 2. Summary graph of oxygen solubility in sodium*

The observed feature of the solubility equations is not accidental. It is a consequence of the well-known compensation law or the Meyer-Neldel Rule [10]. The analysis of impurity solubility in alkali metals and in lead performed by authors [11] has demonstrated a possibility to evaluate (or predict) the mode of existence for the dissolved substances in melts and the entropy difference of solvents (alkali metals and lead). The authors [11] found a correlation between the inverse compensation temperature and the logarithm of molecular mass of the dissolved substance in lithium, sodium, eutectic sodium-potassium alloy and lead:

|  |  |
| --- | --- |
|  | (2) |

In the table presented by them (see Table 2), n = 1 and 103/T\* = 1.79 correspond to the solubility of oxygen in sodium. Meanwhile, a molecular weight of the Na2O compound is M=62, which corresponds to the value of lgM=1.792. This value is exactly within the range of 1.7 ˂ 103/*Т* ˂ 1.8 mentioned above, which corresponds to the temperature range from 558 K to 588 K (285…315°C).

Regularity (2) is also observed for the solubility equations in lead, but at *n* = 0.5. Thus, formula (2) has been confirmed by many real examples.

TABLE 2. CORRELATION BETWEEN COMPENSATION TEMPERATURE *Т*\* AND MOLECULAR MASS OF DISSOLVED SUBSTANCES IN ALKALI METALS [11]

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Solvent | Dissolved element | 103/*Т*\* | lg*M*  of element | lg*M*  of substance | n | Impurity  composition  in melt | Remarks |
| Li | H  N  **O** | 1.50  1.54  **1.475** | 0  1.146  1.204 | 1.502  1.542  **1.475** | 1  1  1 | (LiH)4  Li3N  Li2O | 300-550°С |
| **Na** | H  C  **O**  Si  Pb  Ag | 1.68  1.08  **1.79**  1.45  2.47  2.19 | 0  1.079  1.204  1.447  2.316  2.033 | 1.681  -  **1.792**  -  2.4675  2.187 | 1  1  1  1  1  1 | (NaH)2  C  **Na2O**  Si  Na3,75Pb  Na2Ag |  |
| NaK | H | 1.6 | 0 | 1.603 | 1 | KH |  |
| Pb | **O** | **1.174** | 1.204 | **2.3478** | **0.5** | **PbO** |  |

The logarithm of concentration averaged over 19 known equations of oxygen solubility in sodium at the point 103/*Т*\* = 1.792 is equal to lg*C* = 1.96, RMS is equal to ± 4%, and concentration is *C* = 91.6 ± 9%. The fact that at the temperature equal to the compensation temperature, the solubility of oxygen in sodium corresponds to all the above equations, gives reason to use this point (103/T=1.792; lgC=1.96) as a reference point in the calibration and subsequent periodic verification of a specific plug indicator at a real experimental facility or power plant.

For calibration, it is necessary to determine the oxygen concentration in sodium C1 by means of a sampler-distiller and to measure the plugging temperature *T*1 using the plug indicator in the operating conditions. Two points: (103/*Т* = 1.792; lg*C* = 1.96) and (103/*T*1; lgC1) will specify the solubility equation valid under these conditions.

The samplers-distillers mentioned above are provided in sodium-cooled reactor facilities, but they are rather complicated and cannot be used for on-line control of concentration. However, the sensitivity of the samplers-distillers is about 1 ppm [4], it is sufficient for calibration of the plug indicator from time to time, for example, once per year. The calibrated plug indicator can be used continuously.

1. CONCLUSION

During the lifetime of modern NPPs with sodium (40-60 years), the composition and state of impurities in the coolant change. It results in modification of the solubility equation. Thus, periodic calibration of the plug indicator is required. The proposed calibration method solves this problem.

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