Measurement of High Temperatures Under Irradiation Conditions

THE USE OF MOLYBDENUM-PLATINUM THERMOCOUPLES

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One of the problems in nuclear research is that of measuring temperatures accurately. This problem is of special importance for the high temperature gas cooled Dragon reactor, where it is necessary to measure temperatures up to 1500°C in an environment of graphite and helium.

Base metal thermocouples are only slightly affected by nucleur irradiation and are, therefore, commonly used to measure temperatures up to 1000°C. Rhodium-platinum type thermocouples would be the natural choice for measuring temperatures up to 1800°C and under normal conditions the accuracy, reliability and stability of these thermocouples are well known. However, when used under high neutron flux (1) they suffer from the drawback that rhodium has a very large absorption cross-section for neutrons.

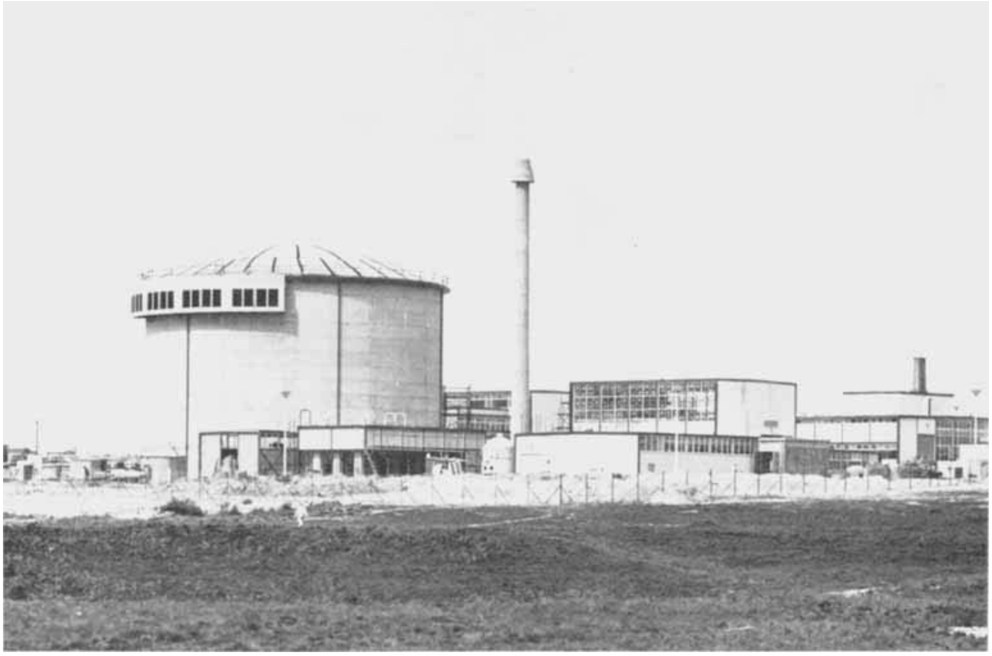
The rhodium atom is capable of capturing neutrons and transforming into an unstable rhodium isotope. This decays to palladium, and hence there is a change in the composition of the rhodium-platinum limb. The output of the thermocouple quickly drifts from the calibrated value since the thermal e.m.f. of a thermocouple is very composition sensitive.

There are two ways of overcoming this problem. The first is to accept the drift as inevitable and to apply a correction factor to the recorded e.m.f. This presupposes that the total dosage is accurately known and sufficient data are available to allow the correction factor to be calculated. For short-term experiments the integrated dosage may be known, but for long-term experiments or for control thermocouples built into the experiment, this amount is almost impossible to determine. It is therefore desirable to use a thermocouple that is unaffected by thermal neutron bombardment.

The choice of metals for this new thermocouple posed many problems. Any two dissimilar metals or alloys may be joined to act as a thermocouple; several metals have low absorption cross-sections, but those that are stable at high temperature are very few in number. Under normal conditions platinum and rhodium-platinum alloys are found

Neutron Absorption Cross-sections for Molybdenum, Rhodium and Platinum and their Effect on the Transmutation Rate

	Absorption cross-section for thermal neutrons	Percentage of atoms converted after an integrated neutron-dose of 10^{21}nvt (thermal)
Molybdenum	2.5×10^{-24} cm^2	0.25
Rhodium	150×10^{-24} cm^2	15
Platinum	8.1×10^{-24} cm^2	0.81



The Dragon Reactor – the high temperature gas-cooled reactor experiment – is being built by twelve European countries, all members of the Organisation for Economic Co-operation and Development (OECD). One of the main objects is to gain operating experience with graphite uranium carbide fuel elements operating at high temperatures for prolonged periods. The outlet temperature of the helium coolant permits steam to be produced with characteristics suitable for modern turbo-alternators

to be the most stable and as platinum has a low absorption cross-section it was chosen as the basis of the new thermocouple. This added the restriction that the other metal must enter into a stable solid solution with platinum and form a ductile alloy capable of being drawn to wire.

Molybdenum was selected (2) as one of the most promising materials that could satisfy these requirements. It can be calculated easily that after an integrated dosage of neutron of 10^{21}nvt—which is typical of the dose received during an irradiation experiment—the percentage of transmuted molybdenum atoms is insignificant and hence that there would be practically no change in the composition of a molybdenum-platinum alloy. The relevant figures are given in the table on the facing page.

Molybdenum is readily soluble in platinum (3) up to 10 per cent by weight and forms a homogeneous alloy. A sensitive check of this latter point is made by drawing an ingot to wire and checking the thermal e.m.f. against pure platinum for samples taken from various parts of the ingot. It is found that the resulting e.m.f.s are all within a very narrow range.

Stability and Compatibility

Assuming that a thermocouple comprising platinum against molybdenum-platinum alloy would be stable under neutron bombardment, the next problem was to examine the factors which affect the long-term stability. Experience with rhodium-platinum thermocouples shows that rhodium contamination of the pure platinum limb can be a problem and that this is minimised if a 1 per cent rhodium-platinum alloy is substituted for pure platinum. It was therefore decided to use a 0.1 per cent molybdenum-platinum instead of pure platinum, although more recent experience has suggested that 1 per cent molybdenum-

Part of a test capsule used in an irradiation experiment in the Dragon Reactor. Fuel is contained in the graphite cylinders and the temperature distribution along the capsule is measured by the metal-clad molybdenum-platinum thermocouples which may be seen protruding through the gaps between the cylinders

platinum may be preferable. The other limb was made of 5 per cent molybdenum-platinum.

The graph on page 125 shows the temperature-e.m.f. relationship for various molybdenum-platinum alloys and it can be seen that the output is almost linear over the range 500 to 1500°C, i.e. the range of direct interest to the designers of the Dragon reactor.

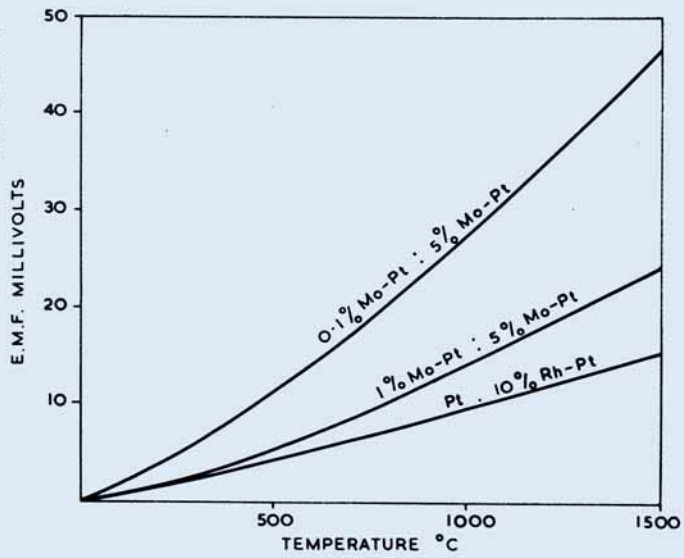
The major disadvantage of these alloys is the tendency to form molybdenum oxide. This, of course, affects the stability. However, in the Dragon reactor high purity, oxygen-free helium circulates and so there is no problem.

A comprehensive series of tests was then undertaken in the Dragon Project laboratories to test the suitability of molybdenum-platinum alloys. Tests were done on bare wire thermocouples insulated with twin-bore alumina beads and thermocouples incorporated into the metal sheathed, mineral insulated form. This latter type was either sheathed entirely with molybdenum-platinum or a composite sheath of part stainless steel and part molybdenum-platinum alloy with magnesia as the powdered insulant.

It was found that molybdenum-platinum was compatible with graphite and helium at high temperatures. This is a most important point since many materials are rapidly corroded under such conditions.

The first test with a bare wire thermocouple showed that after heating for 1000 hours at 1300°C in commercially pure argon (99.95 per cent), a drift of 15 per cent was obtained. The test repeated with the thermocouple inserted in a graphite block produced no drift from calibrated values. The same thermocouple was then heated for 400 hours at 1500°C in vacuum in the presence of graphite and no drift was detected. These tests indicate that even a slightly oxidising atmosphere is

Temperature - e.m.f. relationships for molybdenum-platinum alloy thermocouple compared with the platinum : 10 per cent rhodium-platinum couple



capable of causing substantial drift from calibration and that the presence of graphite inhibits oxidation of the molybdenum and helps to stabilise the thermocouple.

Metal Sheathed Couples

The metal sheathed, mineral insulated form was then subjected to rigorous tests. This type of assembly is the most useful because it is easy to install and requires no extra insulation. The thermocouple is isolated from the sheath and assemblies are readily obtained in sizes down to 0.040 inch outside diameter. The results of the tests were sufficiently encouraging for this type of thermocouple to be used on an experimental basis in the nuclear irradiation programme. It has already been possible to verify that the nuclear stability is very good indeed. Work continues on the examination of the relative metallurgical stability of the 0.1 per cent molybdenum-platinum alloy against 5 per cent molybdenum-platinum, and of the 1 per cent molybdenum-platinum alloy against 5 per cent molybdenum-platinum.

In some of the earlier work the thermocouple wires were found to be contaminated with magnesium after heating at 1250°C for 1000 hours. This may have resulted from the dissociation of the magnesia and experiments are now in hand to test the effect of replacing the present magnesia insulation with alumina.

When the results of all current experiments are available it will be possible to consider the introduction of these thermocouples in the high temperature region of the Dragon reactor. Meanwhile, it is considered that these alloys are most promising materials for measuring temperatures up to about 1700°C under conditions of high neutron flux, provided only that the atmosphere is non-oxidising.

Acknowledgement

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References

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