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Dynamics of hydrogen in sodium in LMFBR secondary circuit

Vivek Nema, D Sujish, B Muralidharan, M Rajan & G Vaidyanathan*
Fast Reactor Technology Group, Indira Gandhi Centre for Atomic Research, Kalpakkam 603 102, India

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Steam generator (SG) of a liquid metal cooled fast breeder reactor (LMFBR) comprises sodium on the shell side transferring heat to water in the tubes. Any crack in the tube will result in the water reacting with sodium producing hydrogen and other corrosive reaction products. This may lead to the damage of the neighbouring tubes. Monitoring of hydrogen concentration in sodium can indicate the initiation of a sodium water reaction (SWR). The hydrogen detectors are based on the diffusion of hydrogen in the sodium through a nickel membrane and measurement of the hydrogen concentration by a mass spectrometer. A numerical model has been developed to estimate the amount of hydrogen concentration in the secondary loop during water/steam leak in SG. The model has been validated based on experiments with hydrogen injection in one loop and actual sodium water reaction in another loop. It has been found that the increase in hydrogen concentration was very well predicted by the code.

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The steam generator (SG) is a critical component of liquid metal cooled fast breeder reactors (LMFBR). Here heat transfer takes place between water (~ 14 MPa) in the tubes and sodium (~ 0.4 MPa) on the shell side of the SG[1]. Due to pre-existing material or weld defects, a crack may develop in the tube leading to sodium water reaction (SWR). The fast detection of sodium water leaks in the steam generator is of paramount importance, in the light of the corrosion and erosion by the reaction products (NaOH, Na_2O, NaH, H_2) leading to damage of the neighboring tubes[2]. One of the methods generally adopted in all LMFBR's to detect a SWR is to measure the hydrogen concentration in the sodium coming out of the steam generator (Fig. 1). In this method, sodium is passed over a nickel membrane through which hydrogen diffuses. The other side of the membrane is connected to a vacuum circuit and mass spectrometer to measure the hydrogen concentration. Experimental testing is usually expensive and time-consuming. A well developed model of the dynamics of hydrogen in the sodium system is a cost-effective tool. It can be used in the design stage to predict the response and optimize the location of the detector. In the operation stage, it is useful to assess the changes affecting the detectors after they are in service by comparing the theoretical estimates and observed values in response to a known hydrogen amount. It can also be used to assess the leak rate based on the response of the detector. Coleman *et al.*[3] developed a one-dimensional model to evaluate the ability of leak detection systems. They did not account for dissolution of hydrogen gas and dissociation of sodium hydroxide. Desmas *et al.*[4] have developed a one-dimensional model and validated the same based on experiments in the MEPHYSTO tests[5]. Their model considered hydrogen dissolution in sodium and

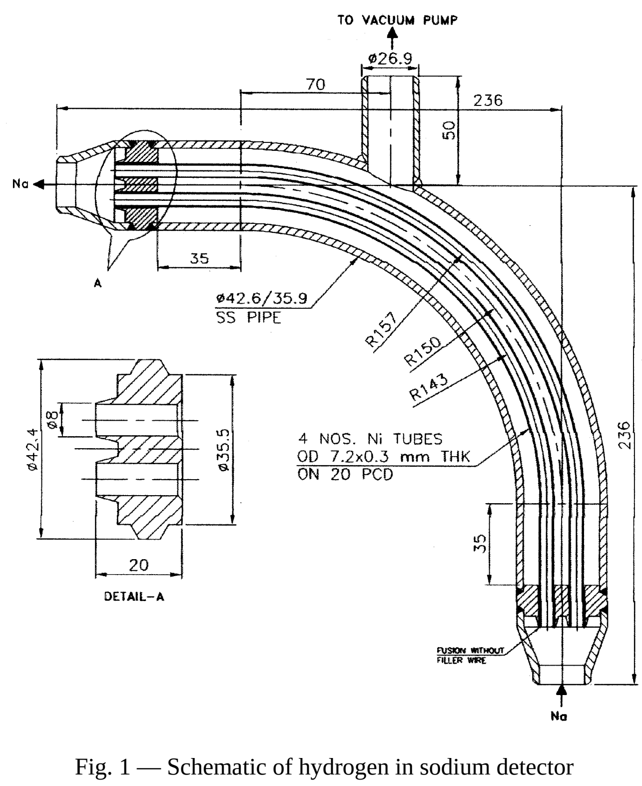

Fig. 1 — Schematic of hydrogen in sodium detector

*For correspondence (E-mail: vaidya@igcar.gov.in) dissociation of sodium hydroxide. Recently, Duan *et al*.[6] have developed a three-dimensional model to study the SWR product transport, where hydrogen dissolution is modelled as a transient point source. The sodium flow field and reaction product concentration fields are obtained by the porosity and distributed resistance approach. They validated the model against experiments in a simple sodium loop. The present paper is an attempt at development of a simple one-dimensional model to predict the hydrogen dynamics and validating the same with tests on two different sodium loops, one with a SG and one without SG. Modelling of the phenomenon involves the hydrogen production, transport, distribution and dissolution in sodium and dissociation of NaOH to produce NaH and Na_2O.

Hydrogen Dissolution

The primary reaction between sodium and water is

$$Na + H_2O \rightarrow NaOH + 0.5H_2$$

This reaction is very fast and can be considered instantaneous. The dissolution of hydrogen bubble in sodium depends on the sodium temperature, initial bubble radius and bubble residence time in sodium. Whittingham[7] has carried out a thermodynamic equilibrium analysis of the phenomena and arrived at the bubble dissolution rate as

$$\mathrm{d}r/\mathrm{d}t = -kT/2700$$

where $\log_{10}k = 6.16 - 4130/T$

Tests conducted based on liquid soda injections in sodium loop MEPHYSTO[5] have indicated an initial hydrogen bubble size of ~1.4 mm. For sodium velocity less than 0.9 m/s, the hydrogen generated is not entrained by the sodium flow and rises to the free cover gas space. Moreover, when the sodium temperature is higher than 420°C the hydrogen formed is dissolved immediately in sodium.

Kinetics of NaOH dissolution

According to the Desmas[4], the dissociation of NaOH is given by

$$\mathrm{d}C_{NaOH}/\mathrm{d}t = -K_{NaOH}\, C_{NaOH}$$

where C_{NaOH} is concentration of NaOH and K_{NaOH} is a constant. The kinetics of dissociation of NaOH has been the subject of much research. In the present model, the experimental results of Kong[5] is used

$$K_{NaOH} = 14.8\, e^{-5725/T}$$

Modelling

The assessment of dynamics of hydrogen requires the equations of transport around the loop. Taking a one-dimensional approach, the transport equation is represented by

$$\mathrm{d}C/\mathrm{d}t + V\,\mathrm{d}C/\mathrm{d}x = S/m$$

where $\mathrm{d}C/\mathrm{d}t$ is rate of change of concentration of NaH or NaOH, $V\mathrm{d}C/\mathrm{d}x$ is the convective term, m the sodium mass in mesh, S is the source term, i.e., mass flow rate of hydrogen injection. C is the concentration of hydrogen in sodium and is a dimensionless number. The assumptions involved are the following: (i) The presence of hydrogen does not alter the sodium flow rate; (ii) The force balance of drag and buoyancy governs motion of hydrogen bubbles. It is assumed that the bubble velocity is same as that of sodium; (iii) Distribution of hydrogen between the sodium and cover gas is important where the free surface vessels like pump tank or expansion tank exist. Hydrogen is highly soluble in sodium if the temperature is greater then 350°C. Since all the injections were made above 350°C of sodium temperature, it is considered that the sodium carries all hydrogen.

A computer code DYNA-H was developed to simulate hydrogen injection tests in sodium loops. In the code, the transport equation for hydrogen is integrated by Euler method. The whole pipe length is divided into a number of meshes such that the mesh length satisfies the Courant criterion.

Brief Description of Sodium Loops

Steam generator test facility (SGTF)

This facility is meant to test a once through steam generator of 5.5 MWT. The schematic sodium circuit of SGTF is given in Fig. 2. The flow rate of sodium in the loop is 55 m^3/h. Sodium at 355°C passes through a fired heater and heated up to 525°C and passes through the surge tank, steam generator and back to the pump suction through buffer tank. A part of the main flow (1 m^3/h) from the SG outlet is passed through hydrogen in sodium detection system (HSD) and in turn it returns to sodium storage tank (SST). Make up of the loop sodium is maintained by pumping sodium from the SST through a purification circuit where the oxides and hydrides of sodium are partly removed. The sensitivity of detection of

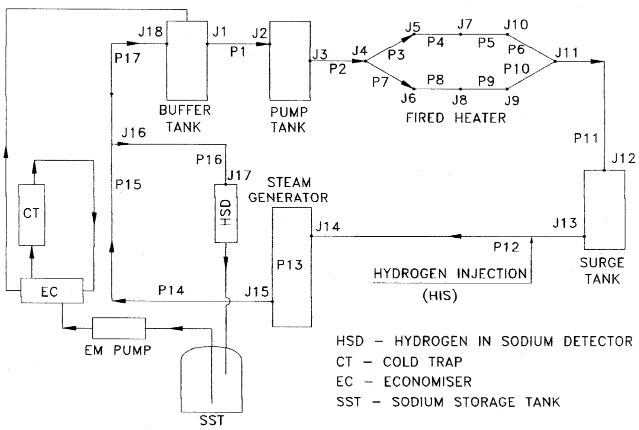

Fig. 2 — Schematic of sodium circuit of SGTF

hydrogen in a sodium system is a function of the operation of purification circuit.

Sodium water reaction test rig (SOWART)

This is an experimental facility designed to study the various aspects of sodium water reaction phenomena. The schematic sodium circuit for SOWART rig is shown in Fig. 3. In SOWART, sodium from storage tank is discharged into the Heater vessel-2 (HV2) by EM pump. The maximum flow rate from EM pump is 20 m^3/h. Sodium from the heater vessel-2 flows through HV1, Expansion tank, NaX and returns to the EM pump. A part of main flow from HV2 outlet is passed through the purification circuit and fed back to the inlet of the EM pump. Hydrogen injection in the loop can be made through Micro leak test section (MLTS) which is provided in between the HV1 and expansion tank. A sodium sample is passed through the hydrogen in sodium detector (HSD) at 1 m^3/h to measure hydrogen concentration.

Calibration in sodium loops

Hydrogen in sodium detection system (HSD)

The HSD is a nickel tube sensor. Sodium flows inside the nickel tube. The exterior of the nickel tube is kept under high vacuum by a sputter ion pump (SIP). The concentration difference causes the hydrogen to diffuse through the nickel tube. The hydrogen thus enters the vacuum side of the meter. The quantity of hydrogen diffused is measured by the difference in SIP current. The quantity of hydrogen in sodium can be obtained using a pre-calibrated curve. The hydrogen leak detection system is calibrated by injecting a known quantity of hydrogen into the loop.

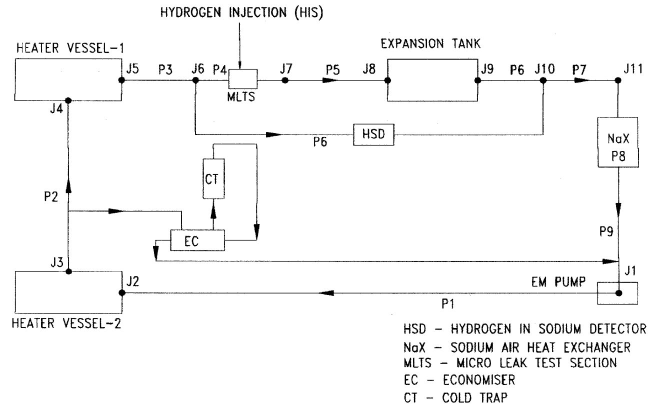

Fig. 3 — Schematic of sodium circuit of SOWART

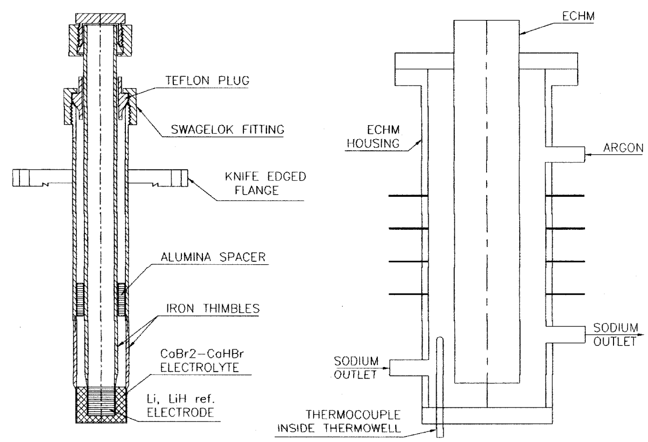

Fig. 4 — Schematic of electrochemical hydrogen meter

Electrochemical hydrogen meter (ECHM)

ECHM sensor contains a $CaBr_2$-CaHBr electrolyte[8]. This electrolyte is filled in the annulus of two thimbles as shown in Fig. 4. Li, LiH filled in the iron thimbles which is located inside acts as a reference electrode. Alumina spacers separate the iron thimbles. The above mentioned sensor is housed in SS pipe through which the sample sodium flows. The sodium containing hydrogen with reference electrode and electrolyte forms an electrolytic cell.

Testing of HSD in SGTF

To detect the sodium water reaction (SWR), hydrogen in sodium detection system (HSD) is provided in the sodium circuit. The sodium line from SG outlet is by passed through HSD for sampling. Sodium flow rate through the HSD is maintained at 1 m^3/h. Water/steam leak in sodium can be detected by measuring the increased concentration of hydrogen in sodium. Hydrogen detection circuit consists of electrochemical hydrogen meter (ECHM) and diffusion type nickel detector in series. To calibrate the diffusion type nickel detector, a known quantity of hydrogen was injected into the system through hydrogen injection system (HIS). Three hydrogen injections were carried out and the results obtained during the experiment are shown in Fig. 5. The hydrogen concentration is constant at background level and it increases as the slug of hydrogen reaches to the detector location (HSD). Later the detector sees hydrogen after one circulation time of sodium around the loop (about 14 min) and the magnitude of concentration is decreasing with time due to the dilution of hydrogen in remaining sodium of the loop. With second and third injections the detector response is as expected.

Testing of HSD in SOWART

HSD system similar to SGTF has been provided to detect the sodium water reaction (SWR) in SOWART. Water/steam leak in sodium can be detected by measuring the increased concentration of hydrogen in sodium. Three injections of hydrogen concentrations 40 ppb, 40 ppb and 100 ppb were carried out the results obtained during the experiment are shown in Fig. 6.

Results and Discussion

SGTF

Three hydrogen injections were simulated using the Dyna-H code. The quantity and rate of hydrogen injected in each injection is given as follows.

First injection	0.38 g	0.63 mg/s
Second injection	0.62 g	1.0 mg/s
Third injection	0.94 g	1.5 mg/s

The schematic flow diagram used for simulation of hydrogen injection in SGTF is shown in Fig. 2. It is assumed that sodium flows through buffer tank, pump tank, fired heater, surge tank, steam generator and back to the buffer tank in a closed loop. The sodium storage tank has been omitted because the sodium flow rate to the storage tank from the HSD system is very low (1 m^3/h) when compared to the flow in the main loop. The change in concentration due to the mixing of 1 m^3/h sodium flow in storage tank can be neglected. The purification circuit like cold trap (CT), plugging indicator are also not taken into consideration because the cold trap was isolated

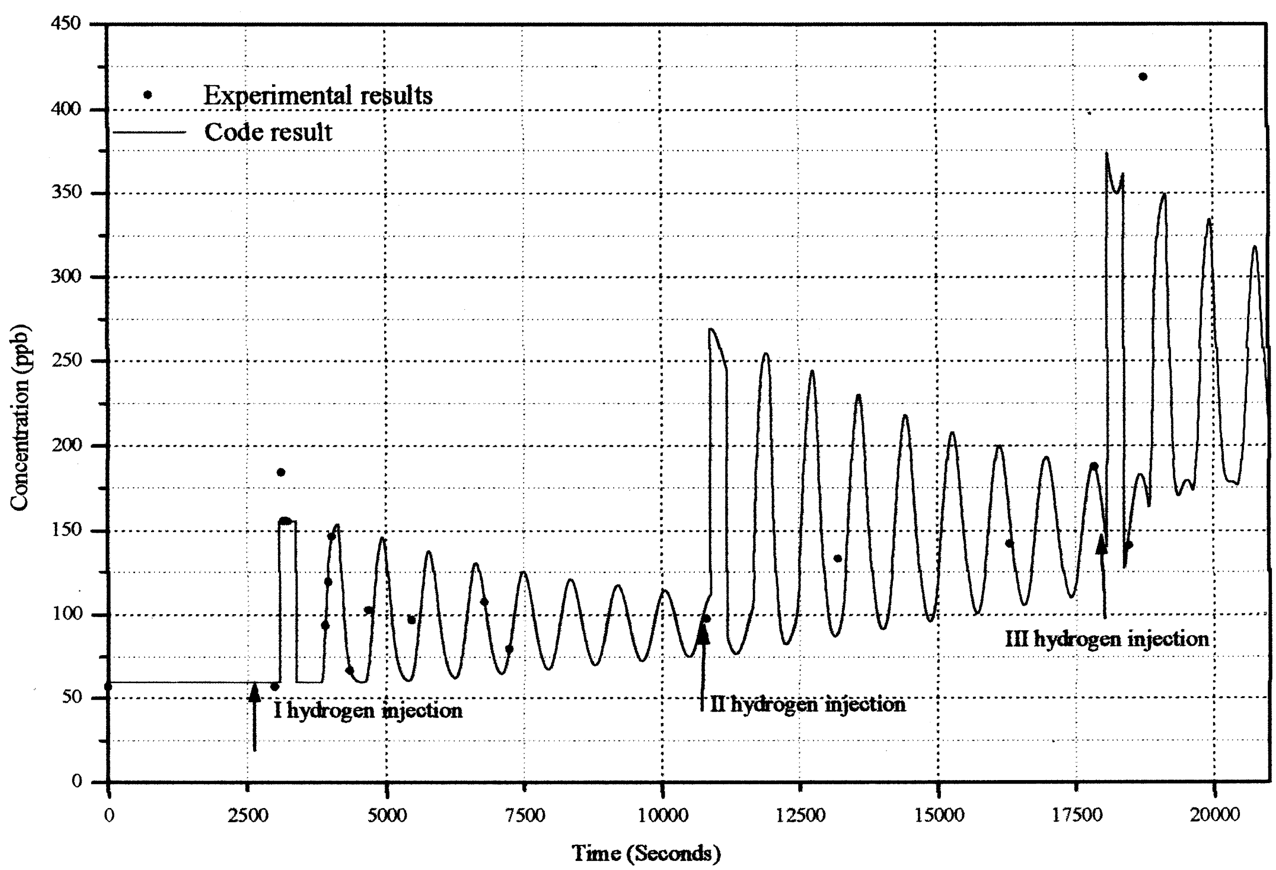

Fig. 5 — Comparison of hydrogen injection in SGTF with code

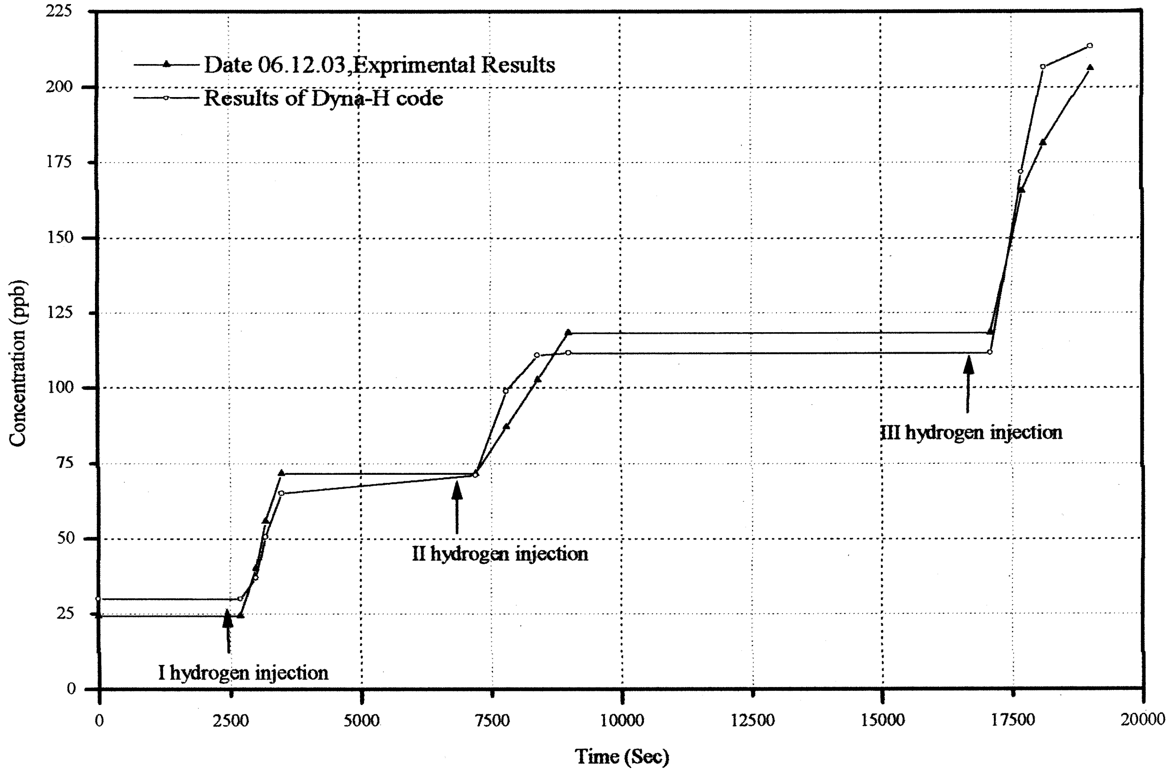

Fig. 6 — Comparison of hydrogen injection in SOWART with code

during calibration. Figure 5 shows comparison of the experimental and theoretical results. It can be seen that the initial fluctuations of hydrogen concentration at the detector location are very well predicted by the code.

SOWART

Three injections were carried out in SOWART and the same were simulated using Dyna-H code. The schematic flow diagram used for simulation of hydrogen injection in SOWART rig is shown in Fig. 3. The effect of sodium storage tank on the result is negligible and hence storage tank has not been modelled here as well. Since the cold trap was isolated during calibration, the purification circuit is omitted. It can be noticed (Fig. 6) that experimental results are very well predicted by the code.

Conclusions

A one-dimensional model of hydrogen dynamics has been developed. This model has been used to determine the hydrogen concentrations at the detector location for two different sodium rigs after injection of hydrogen for calibration. It is seen that there is a good comparison between the predictions and experiments. It is clear that such models are adequate from the point of operating personnel to know the leak rate. It is expected that such models could be introduced in the simulator models for operator training.

In this work, it is assumed that all the hydrogen generated is dissolved in sodium and nothing escapes to cover gas space. This assumption is valid for small water/steam leak case and for sodium temperature above say 350°C. As a future work it is planned to simulate the case where in some quantity of hydrogen escapes to cover gas space due to either operation at low sodium temperature (<300°C) or medium leak of water/steam. Also impact of the operation of purification circuit on the hydrogen dynamics needs to be modeled.

References

1 Muller R A & Schnauder H, *Atomic Energy Rev,* 13 (1975) 215-323

2 Dumm K, *Leak detection in LMFBR SG during operation,* IAEA/IWGFR Specialist meet, Dimitrovgrad, USSR, (1978), 29-40

3 Coleman J H & Hosler A H, *Sodium-water reaction analysis of the Babcock & Wilcox prototype steam generator in the sodium components test installation,* APDA-265, (1970)

4 Desmas T, Kong N, Maupre J P, Schindler P & Blanc D, *Hydrogen Detection System Leak Response Codes,* IAEA, IWGFR/78, Proc Specialists Meeting on SG failure and

Failure Propagation experience, Air-en-provence, France, Sept. 26-28,1990, 205-216.

5 Kong N, Fang Z, Desmas T, Lebris A & Hobbes P, *Experimental Tests in Support of Hydrogen Detection Code Development,* Int Conf Fast Reactors and Related Fuel Cycles, Kyoto, Japan, Oct. 28-Nov. 1,1991, 3, P3-19-1.

6 Riqiang Duan, Zhou Wang, Xianyong Yang & Rui Lui, *J Nucl Sci & Technol*, 38(7) (2001) 527-532

7 Whittingham A C, *J Nucl Mater,* 60 (1976) 119-131

8 Kitheri J, Sujatha K, Nagaraj S, Mahendran K H, Sridharan R, Periaswami G & Gnanasekaran T, *J Nucl Mater,* 344 (2005) 285-290