ABSTRACT

Permeation of gases at low partial pressure is of profound interest to the nuclear power industry. The present thesis on gas permeation comprises of two parts, and each part is complete with its own bibliography.

In **Part-I** of the thesis, the generalized field equations for permeation in presence of traps of various kinds, reversible and/or irreversible, at low as well as high trap coverage, with or without surface impedances were solved by a mass balance integral method to yield the closed form functional relationships of permeability as a function of time and trapping parameters. The permeation rates predicted by these expressions agree well with the present finite difference solution, as well as, with the output of existing numerical and series solutions. Subsequently, powerful computer softwares were developed for determining the trapping parameters from the experimental permeation data by a curvilinear regression on these functional relationships. Several data of the hydrogen permeability in the literature were analyzed with the help of the present program to investigate the nature of trapping. A detailed parametric study on the basis of the present model established several characteristic features of permeation under the stipulated conditions. For instance, the time for the onset of any significant permeation flux is insensitive to the trapping parameters, if the traps are of one kind and the trap coverage is low, while it is not so for mixed traps at high trap coverage. In the latter case, the pseudo-steady state flux seems to be directly related with the average capture strength of the traps. Moreover, for small coverage of any one kind of traps, the so called double plateau characteristics appears to be superfluous.

In **Part-II** of this thesis, the development of a modified highly sensitive ^{222}Rn permeation measuring equipment capable of measuring Rn flux on atomic scale (down to ~ 5000 atoms m^{-2} s^{-1} or less) has been reported. Permeation rate of ^{222}Rn (N_{Rn}) in high purity annealed Pb and Au, as well as, in cold worked Au was measured at different temperatures (e.g., 30 - 180 C) in course of prolong (≥135 h) exposure to the permeating ^{222}Rn gas by means of this equipment to reveal various facets of this inert gas permeation in metals at low partial pressure (~ 10^{-7}).

Virgin annealed Au was impermeable to Rn up to 150 C ; but it became permeable after being subjected to one thermal cycle of negative magnitude (TCNM). On the other hand, virgin cold worked Au was highly permeable even at room temperature. Prolong isothermal holding resulted the permeation rate to reach a plateau in annealed Pb and Au pre-annealed at 700 C for 1h (Au700). Similar isothermal treatments produced well defined permeation peaks in cold worked Au, as well as, in Au pre-annealed at 850 C for 1h (Au850). The activation energy of permeation (Q_P) in cold worked Au (18.0 to 22.5 kJ/mol) was appreciably smaller than that in Au700 (33.2 to 35.6 kJ/mol) ; but the N_{Rn} in the former was about two orders of magnitude higher than that in the Au700. In Au850, though the Q_P values are almost the same as that of Au700, the maximum attainable N_{Rn} was only ~ 0.04 times of that in Au700 of identical thickness at the same permeation temperature, T_P (e.g., 100 C). During isothermal treatment, the Q_P in Pb (8.2 kJ/mol) was substantially smaller than the same in Au. These observations have been interpreted in terms of the surface roughness of the specimens, as well as, the capture of vacancies by diffusing ^{222}Rn atoms and the structural changes in the diffusion paths (e.g., grain boundaries) by deformation induced defects.

The effect of the number of TCNM (*Z*) on the permeability in all the above mentioned specimens was studied at T_P = 150 C, after prolong equilibration at the same temperature (i.e., 150 C). In all the cases the permeability enhanced remarkably with Z to reach a peak value $N_{Rn(peak)}$, following which there was a gradual reduction of N_{Rn} with Z. The level of $N_{Rn(peak)}$ increased with the amplitude of TCNM. In case of Pb, the Q_P decreased to an insignificant level of 1.2 kJ/mol due to the preceding TCNM, while in case of Au700 or Au850 similar treatment diminished Q_P to a level comparable to that in the virgin cold worked Au. In the case of Pb the results may be explained in terms of collection of vacancies by large ^{222}Rn atoms during temperature variation in course of the TCNM, that may lead to the formation of small "channels" of atomic dimension. In annealed Au the enhancement in free volume of the permeation paths caused by TCNM may be responsible for the remarkable increase in permeability. It appears, that the TCNM may act as a 'vacancy pump' and the structural defects introduced by it tend to annihilate during subsequent prolong isothermal holding at ambient temperatures. In fact, the loss of permeability caused by the low temperature (e.g., 50 C) isothermal holding of virgin cold worked Au could be partially regained by TCNM, which possibly emphasizes the role of excess vacancies present in the effective permeation paths like grain boundaries.