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An Improved Modelling Approach of Moisture Absorption in Adhesive Joints Using the Finite Element Method

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An Improved Modelling Approach of Moisture Absorption in Adhesive Joints Using the Finite Element Method

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ABSTRACT

This document presents an improved scheme that has been developed to enable the assessment of the moisture uptake in adhesive joints utilising the finite element method. A transient finite element procedure has been applied to accurately predict the moisture concentration within the adhesive layer. This approach can accommodate irregular joint geometries in three dimensions. It offers substantial time savings compared to the previously used analytical approach since the nodal concentration results can be directly transferred to the global FE model and readily linked with the moisture-dependent mechanical properties. Comparison of the finite element and analytical results for a one-dimensional diffusion problem showed excellent correlation.

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Approved on behalf of Managing Director, NPL, by Dr C Lea,
Head of Centre for Materials and Technology.

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ILLUSTRATIONS

1. INTRODUCTION

Predictive modelling techniques have been increasingly used to provide information on the environmental effects in adhesively bonded joints. Special-purpose analytical formulations have been developed to determine the moisture distribution and moisture content as a function of time for simple joint geometries. The effect of moisture migration on the mechanical properties of adhesives has been recently investigated by means of the finite element method (FEM). The subject is a very complicated one, primarily due to the coupled mechanical diffusion response that influences the joint behaviour.

Current research at the National Physical Laboratory (NPL) includes the use of commercial FE software to model the redistribution of stress and strain in bonded joints due to moisture ingress. The bulk diffusion is considered to be the primary transport process and interfacial diffusion is neglected. Consequently, weakening of the joint due to moisture absorption is assumed to occur through plasticisation of the adhesive and the failure is cohesive in nature. The modelling procedure consists of finding the temporal and spatial distribution of moisture within the adhesive layer and modelling the mechanical-diffusion interaction. The experimental data required to carry out the coupled mechanical-diffusion analysis are the moisture-dependent mechanical properties of the adhesive and the relevant diffusion parameters.

The work presented in this document is concerned with the prediction of the moisture distribution using the finite element method. Previously, one-dimensional analytical formulations were used to compute the moisture concentration at nodal co-ordinates and inclusion in the finite element model was performed manually. A new approach incorporating a sequentially coupled mechanical-diffusion finite element analysis is discussed in this report.

2. ANALYTICAL FORMULATION

Analytical expressions for the moisture distribution as a function of time of homogenous materials exposed on one or both sides to water are presented by Shen and Springer [1]. The problem is pictured in Figure 1, where the plate is taken to be infinitely long in the *y*- and *z*-directions. The moisture content inside the plate varies only in the *x*-direction, i.e. the problem is one-dimensional. Initially the moisture concentration c_i inside the plate is uniform. The plate is suddenly exposed to a moist environment and the exposed faces reach instantaneously the moisture concentration c_a which remains constant.

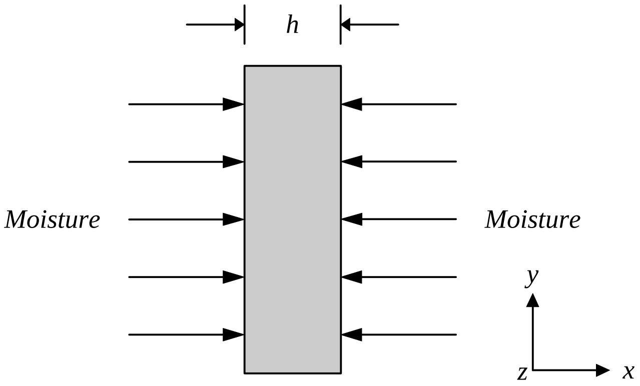

Figure 1 *- Graphical representation of one-dimensional diffusion problem*

Fick's law [2]:

$$\frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2} \tag{1}$$

where D_x is the diffusivity of the material and the boundary conditions are:

$$c = c_i \qquad 0 < x < h \qquad t \leq 0 \tag{2a}$$

$$c = c_a \qquad x = 0; x = h \qquad t > 0 \tag{2b}$$

It has been observed [3] that the diffusivity changes very little with moisture content and thus the solution to equations (1) and (2) is given by [4]:

$$\frac{c(t)-c_i}{c_a-c_i} = 1-\frac{4}{\pi}\sum_{j=0}^{N}\frac{1}{(2j+1)}\sin\frac{(2j+1)\pi x}{h}\exp\left[-\frac{(2j+1)^2\pi^2 D_x t}{h^2}\right] \tag{3}$$

where N is the number of summation terms and *c(t)* is the instantaneous concentration.

3. FINITE ELEMENT DIFFUSION ANALYSIS

3.1. FINITE ELEMENT FORMULATION AND MODELLING APPROACH

The commercial finite element code ABAQUS is used to model the mass diffusion process, with the governing equations for mass diffusion in ABAQUS being an extension of Fick's law. The model allows for non-uniform solubility of the diffusing substance in the base material and for mass diffusion driven by gradients of temperature and pressure [5]. The basic solution variable is the normalised concentration

$$j = {}^{c}\!/_{s}$$

where *c* is the mass concentration of the diffusing material and *s* is its solubility in the base material. This type of definition enables *j* to be continuous across the interface between different materials.

In the joints considered wherein, mass diffusion through the metal substrates is assumed to be negligible and thus the diffusion model does not include dissimilar materials that share nodes. Since no material interfaces are present, the solubility of the adhesive is defined as unity so that concentration and normalised concentration are equivalent.

An important issue in transient diffusion problems with second-order elements is the choice of initial time step. Because there is a relationship between the minimum time step and the spatial element size, spurious oscillations may occur in the solution when the initial time step is smaller than a certain value. ABAQUS provides no check on the initial time increment defined so the user must ensure that the given value is appropriate. A suggested criterion is [5]:

$$\Delta t \geq \frac{\Delta l^2}{6D}$$

where *D* is the diffusivity and *Dl* is a characteristic element size.

Because the rate of change of normalised concentration varies widely during the analysis it is recommended that automatic time incrementation is used for transient analysis. This enables time increments to change in order to maintain accuracy in the time integration. The accuracy is controlled by specifying the maximum normalised concentration change allowed at any node during an increment. Finally, transient analysis can be terminated by completing a specified time period or by reaching steady-state conditions. Steady state is reached when all normalised concentrations change are less than a user-defined rate.

3.2. COMPARISON WITH ONE-DIMENSIONAL ANALYTICAL FORMULATION

In order to verify the mass diffusion capability in ABAQUS, a one-dimensional problem is analysed. A typical single-lap joint configuration has been considered where only half of the overlap has been modelled due to symmetry. A schematic representation of the lap joint overlap region is shown in Figure 2. A diffusion coefficient D_x of $1.1\times10^{-12}\text{m}^2\cdot\text{s}^{-1}$ had been used as a representative value for structural adhesives [6]. Since the problem is one-dimensional, the only gradient is along the adhesive, hence, a plane mesh with only one element in the *y*-direction is used. The mesh is refined near the edge of the adhesive where high concentration gradients are expected.

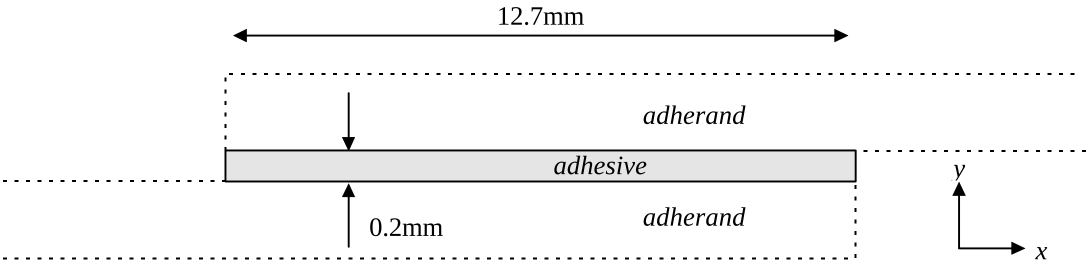

Figure 2 *- Graphical representation of joint overlap region*

The moisture concentration at the exposed edge of the adhesive was specified instantaneously at the start of the step via the concentration degree of freedom at the equivalent nodes of the mesh. The transient diffusion process was modelled for 278hrs. The analysis was performed in three steps in order to compare results at specific times during the transient: 10hrs, 100hrs and 278hrs. The variation of concentration with time determined by the FEM is depicted in Figure 3. Comparison with the analytical results of Shen and Springer [1] is excellent, as shown in Figure 4. This case study was also analysed by Crocombe [6] using both approximate and finite elements methods with the results being in very good agreement with the results from the ABAQUS code. FEM results were identical for first- and second-order elements, while similar results were obtained with much coarser meshes.

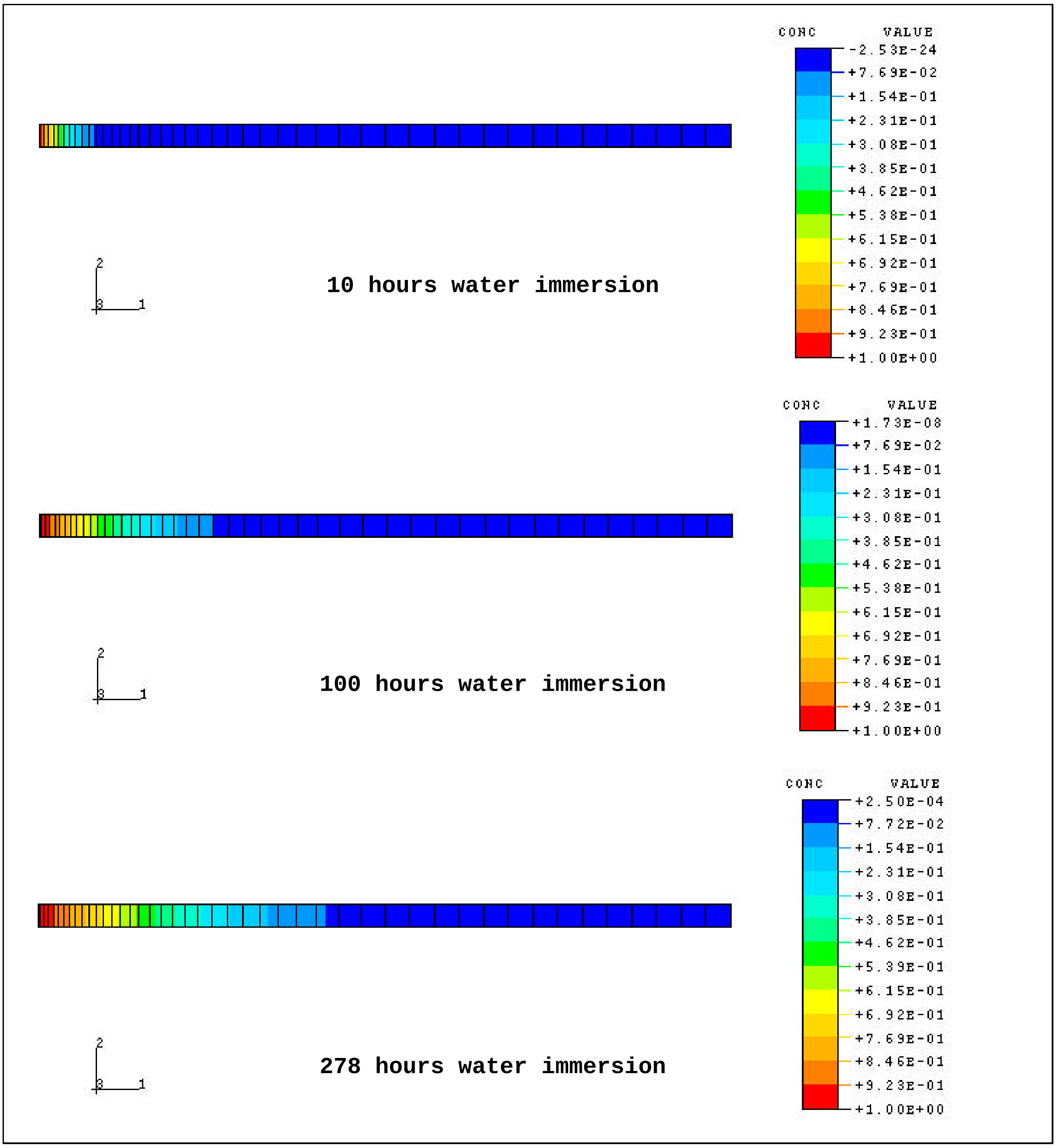

Figure 3 - *Contour plots of normalised moisture concentration along adhesive overlap computed by the FEM*

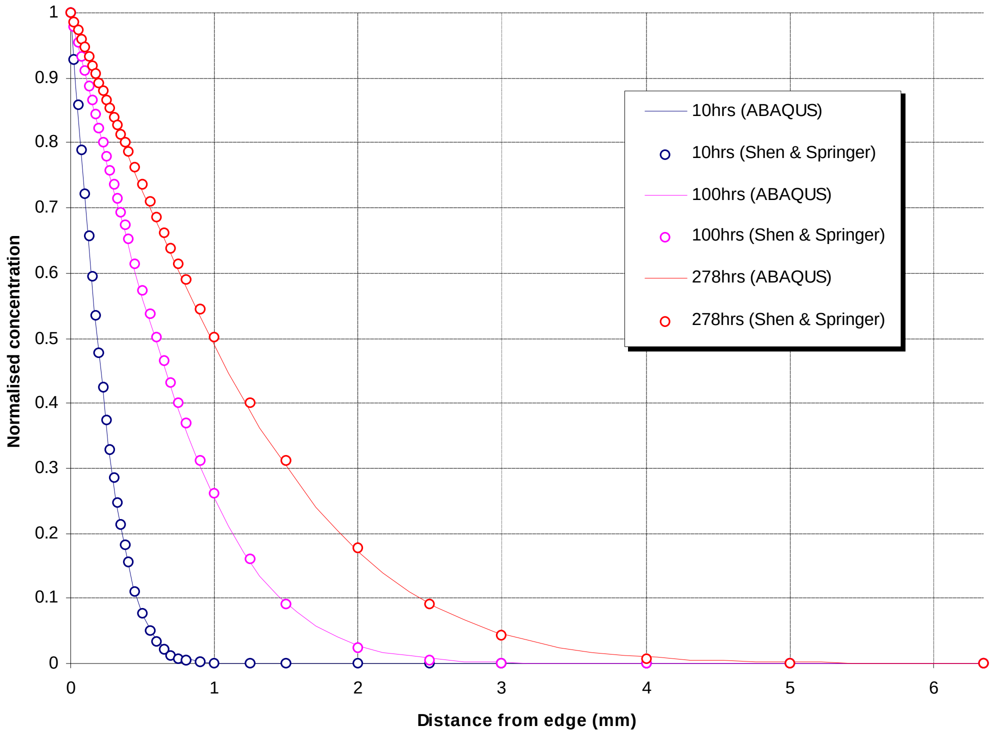

Figure 4 *- Moisture distribution along adhesive overlap*

4. MASS DIFFUSION IN JOINTS WITH IRREGULAR GEOMETRY

Simple expressions for moisture diffusion in adhesive layers do exist. However, these are generally based on linear Fickian analysis of thin rectangular strips. In cases where the geometry is irregular and/or the problem is no longer one-dimensional, a simple analytical solution is not available.

Two irregular geometries, a T-peel joint and a perforated lap joint, are considered in order to assess the efficiency of the analytical solution and illustrate the usefulness of the finite element method. In the case of the T-peel joint the adhesive fillet occupies either half or all of the space between the metal substrates, as depicted in Figure 5. Equation (3) was used to compute the moisture distribution at the nodal co-ordinates within the adhesive in the T-peel joint with 50% fillet, assuming the only concentration gradient is along the bond length. Figure 6 depicts the analytical and finite element solutions for a time period of 288 hours using a diffusion coefficient of $6.7\times10^{-12} m^2 \cdot s^{-1}$ [7]. The boundary conditions assigned were that the concentration reaches its maximum value instantaneously on the exposed adhesive faces and that the concentration gradient normal to the other edges is zero.

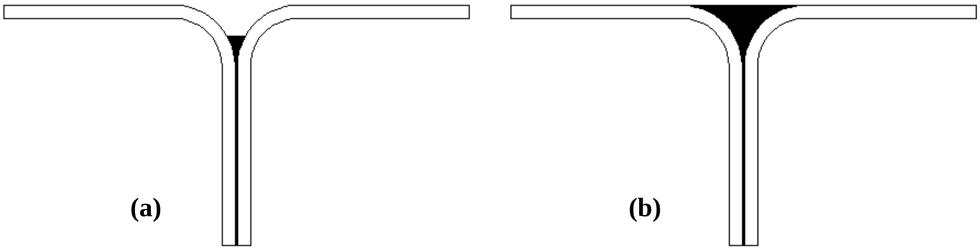

Figure 5 **-** *Schematic of the T-peel joint with (a) 50% resin fillet and (b) 100% resin fillet*

It can be clearly seen in Figure 6, that the one-dimensional solution under-predicts the moisture absorption at the wider end of the adhesive. The computed concentration distribution near the thin end of the layer is almost identical for both analyses as the transport process is essentially one-dimensional. It should be noted that the results from the finite element diffusion model can be directly transferred to the overall finite element model for a subsequent stress analysis.

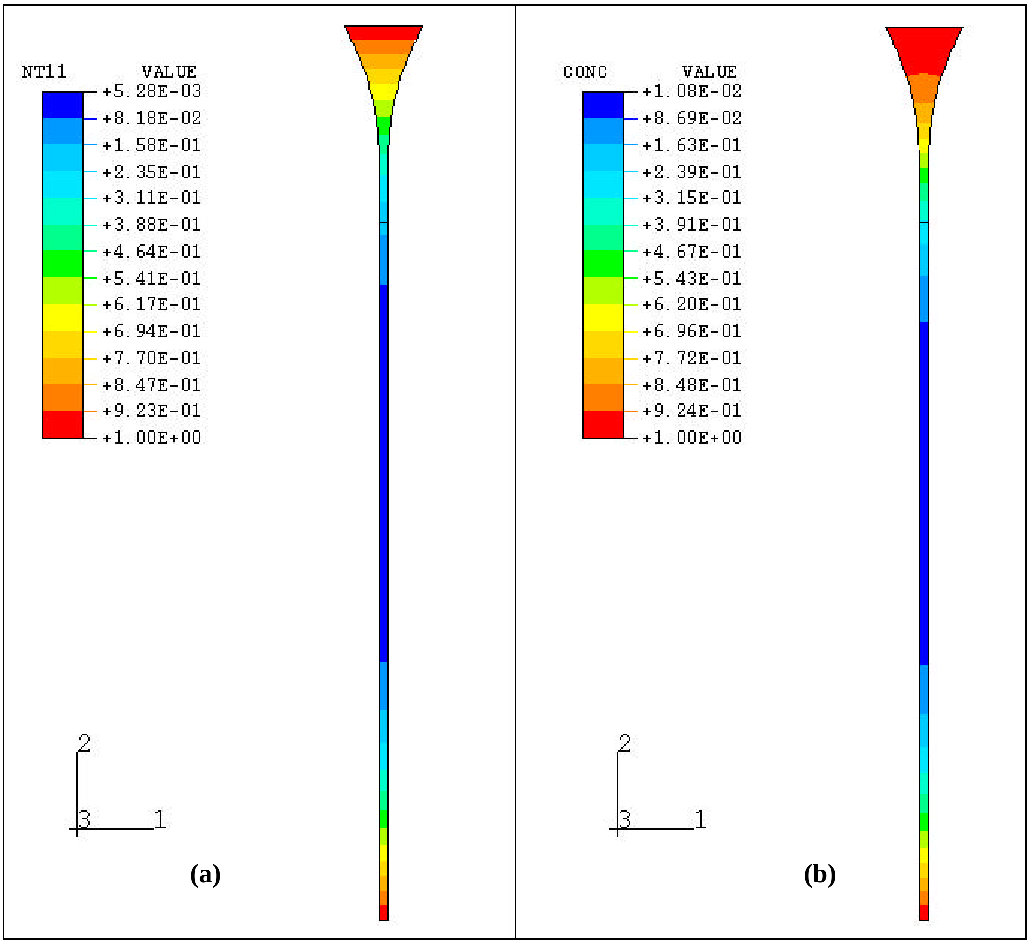

Figure 6 **-** *Contour plot of normalised moisture concentration for T-peel joint*

(a) results from analytical solution mapped on the FE mesh (b) finite element solution

Numerical predictions for the moisture concentration between joints with 50% and 100% resin fillet were also compared. It can be seen from Figure 7, that the moisture concentration at the edge of the overlap is 41% lower when the T-peel joint has a full resin fillet. Results from the transient finite element analyses of the T-peel joint with 50% and 100% resin fillet are contained in Appendices B and C, respectively.

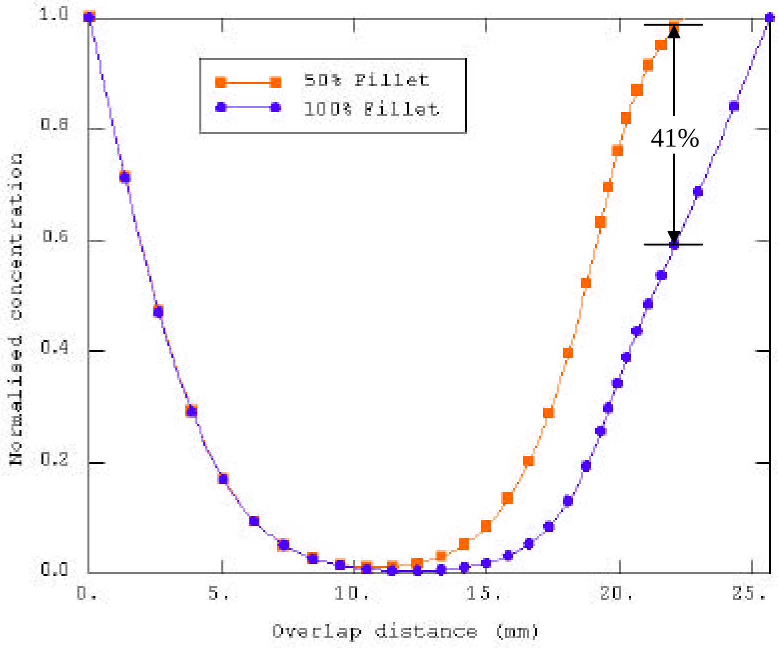

Figure 7 *- Effect of resin fillet on moisture distribution in T-peel joint*

The second case study considers a single lap joint where the overlap area has been perforated in order to increase moisture ingress. Three equally spaced circular holes have been created along the width of the joint necessitating the use of a three-dimensional model for the mechanical response to be instigated. A symmetric finite element model of the adhesive layer has been constructed for the diffusion modelling part of the analysis. Different mesh sizes were also investigated based on stress analysis experience. Moisture concentration results varied only slightly between coarse- and fine-meshed models. This is shown in Figures 8 and 9. Finite element results at different time intervals are presented in Appendix D. A primary observation is that a converged solution in the region of the adhesive in a diffusion problem is, generally, achieved with a coarser mesh than in an equivalent stress analysis for the same joint geometry. Therefore, in a sequentially coupled diffusion-mechanical analysis the mesh density will be generally determined based on the accuracy requirements of the mechanical analysis.

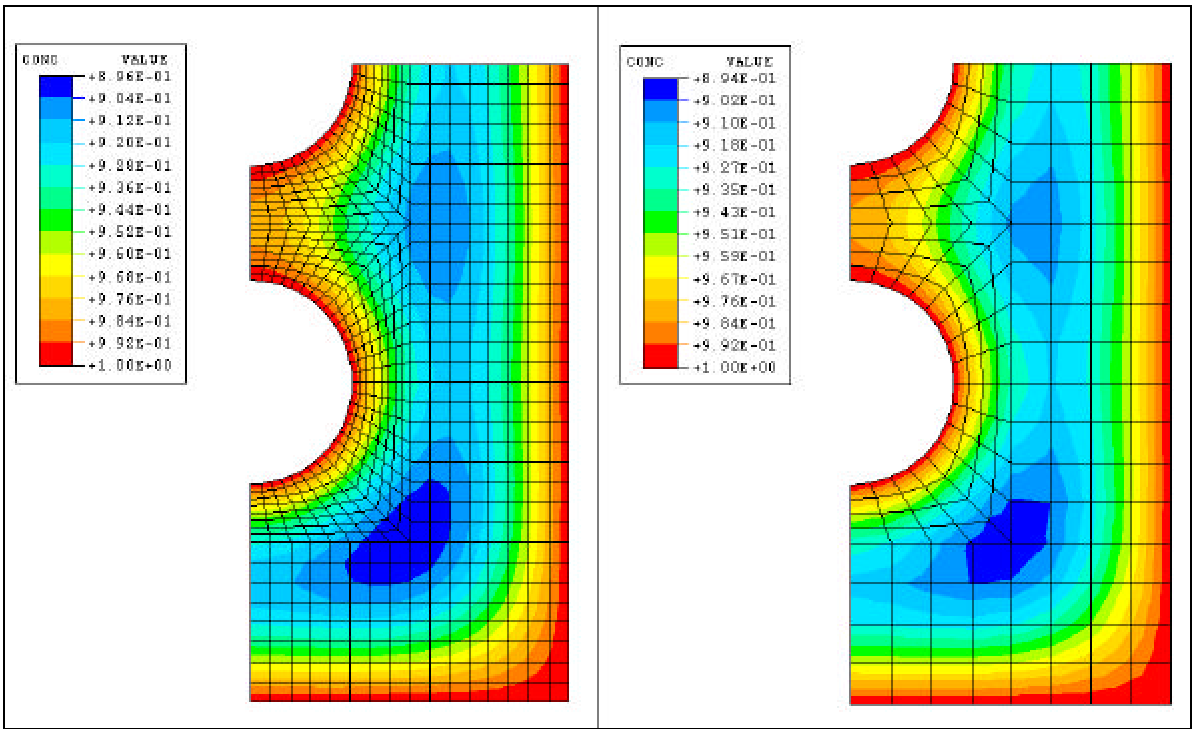

Figure 8 - *Contour plot of normalised moisture concentration for perforated lap joint*

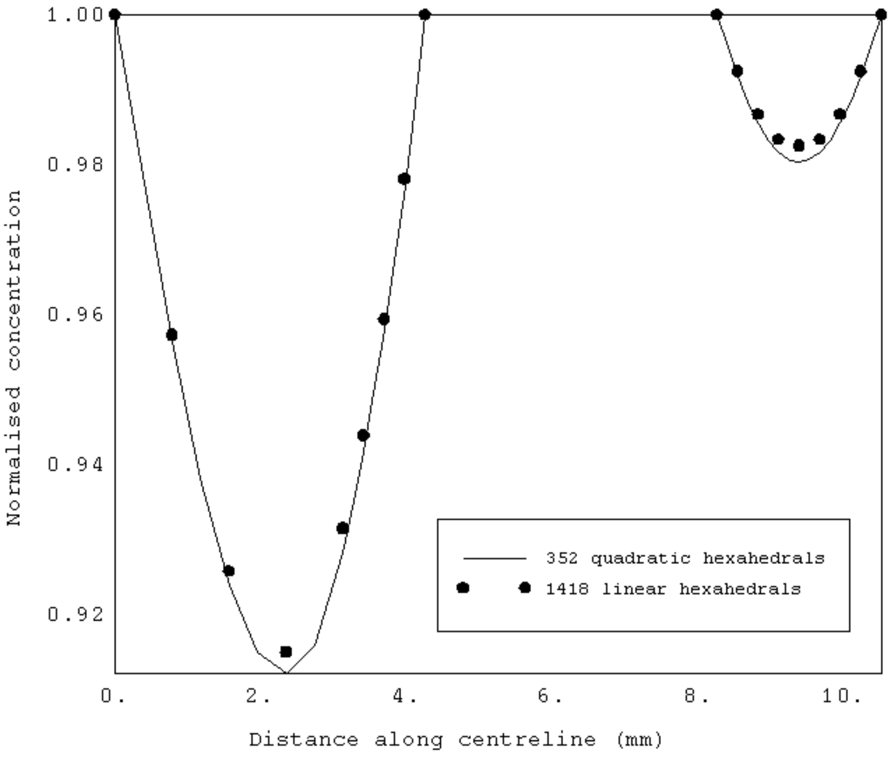

Figure 9 - *Moisture distribution along the centreline of perforated lap joint*

5. CONCLUSIONS

A general scheme has been presented that forms the basis of the diffusion modelling in the sequentially coupled diffusion-mechanical analyses of adhesive joints. Previously, determination of the moisture distribution within the adhesive was determined using analytical expressions and the association with the finite element mesh was performed manually. This resulted in a tedious and time consuming model-building process, particularly when large models were involved. Moreover, the analytical solutions are a one dimensional approximation of the mass diffusion problem and induce errors when the geometry is irregular. The proposed scheme incorporates a transient finite element technique, sequentially coupled with a mechanical analysis. This results in more accurate representations of the moisture concentration field within the adhesive layer, allowing for irregular joint geometries in three dimensions. It eliminates the need for the manual association of nodal concentration values. Finite element and analytical results for a one-dimensional diffusion problem were in excellent agreement.

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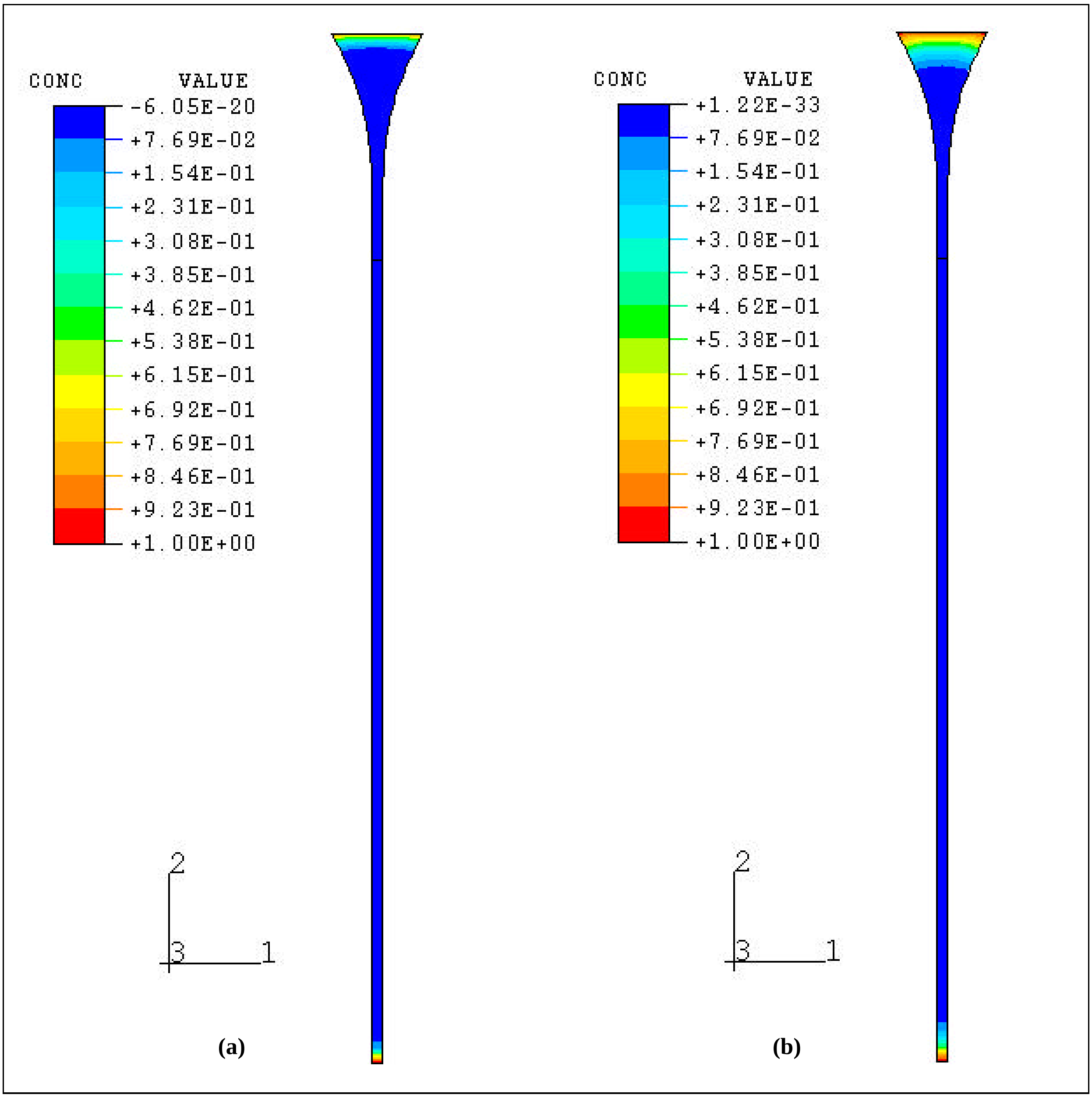
APPENDIX A:

ABAQUS LISTING FOR ONE-DIMENSIONAL PROBLEM

```
*HEADING
 ONE-DIMENSIONAL DIFFUSION EXAMPLE
 PREPARED BY G.HINOPOULOS
 CMMT, NATIONAL PHYSICAL LABORATORY
*RESTART,WRITE,FREQUENCY=1
*PREPRINT, ECHO=NO, MODEL=NO, HISTORY=YES
*NODE, NSET=NODEALL, SYSTEM=R
    1,     0.0000000,     0.0000000,     0.0000000
    2,     0.0000000,     0.2000000,     0.0000000
    3, 0.3759536E-01,     0.0000000,     0.0000000

... more node definitions...

   88,      6.123214,     0.2000000,     0.0000000
   89,      6.350000,     0.0000000,     0.0000000
   90,      6.350000,     0.2000000,     0.0000000
*ELEMENT, TYPE=DC2D4, ELSET=ADHESIVE
    1    1    3    4    2
    2    3    5    6    4

... more element definitions...

   43   85   87   88   86
   44   87   89   90   88
*NSET, NSET=LHEND
 1, 2
*SOLID SECTION,MATERIAL=POLY,ELSET=ADHESIVE
 1.0E+00
*MATERIAL,NAME=POLY
*SOLUBILITY
 1.0E+00
*DIFFUSIVITY, TYPE=ISO
 3.96E-03
*INITIAL CONDITIONS,TYPE=CONCENTRATION
 LHEND, 1.0E+00
*STEP, AMPLITUDE=STEP, INC=100, UNSYMM=YES
 PHASE 1: 10 HOURS
*MASS DIFFUSION, DCMAX=0.10E+00, END=SS
 1.0E-01, 10.0E+00, 1.0E-10, 100.0E+00, 1.0E-08
*BOUNDARY
 LHEND,  11,11, 1.0E+00
*EL PRINT,FREQUENCY=999,POSITION=AVERAGED AT NODES,ELSET=ADHESIVE
 CONC
*END STEP
*STEP, AMPLITUDE=STEP, INC=100, UNSYMM=YES
 PHASE 3: 100 HOURS
*MASS DIFFUSION, DCMAX=0.10E+00, END=SS
 1.0E-01, 90.0E+00, 1.0E-10, 100.0E+00, 1.0E-08
*BOUNDARY
 LHEND,  11,11, 1.0E+00
*EL PRINT,FREQUENCY=999,POSITION=AVERAGED AT NODES,ELSET=ADHESIVE
 CONC
*END STEP
*STEP, AMPLITUDE=STEP, INC=100, UNSYMM=YES
 PHASE 3: 278 HOURS
*MASS DIFFUSION, DCMAX=0.10E+00, END=SS
 1.0E-01, 178.0E+00, 1.0E-10, 100.0E+00, 1.0E-08
*BOUNDARY
 LHEND,  11,11, 1.0E+00
*EL PRINT,FREQUENCY=999,POSITION=AVERAGED AT NODES,ELSET=ADHESIVE
 CONC
*END STEP
```

APPENDIX B:

RESULTS FROM TRANSIENT DIFFUSION ANALYSIS OF T-PEEL JOINT (50% FILLET)



(a) **(b)**

Figure B.1 - *Contour plot of normalised moisture concentration for T-peel joint after:*

(a) 0.695 hours and (b) 3.71 hours

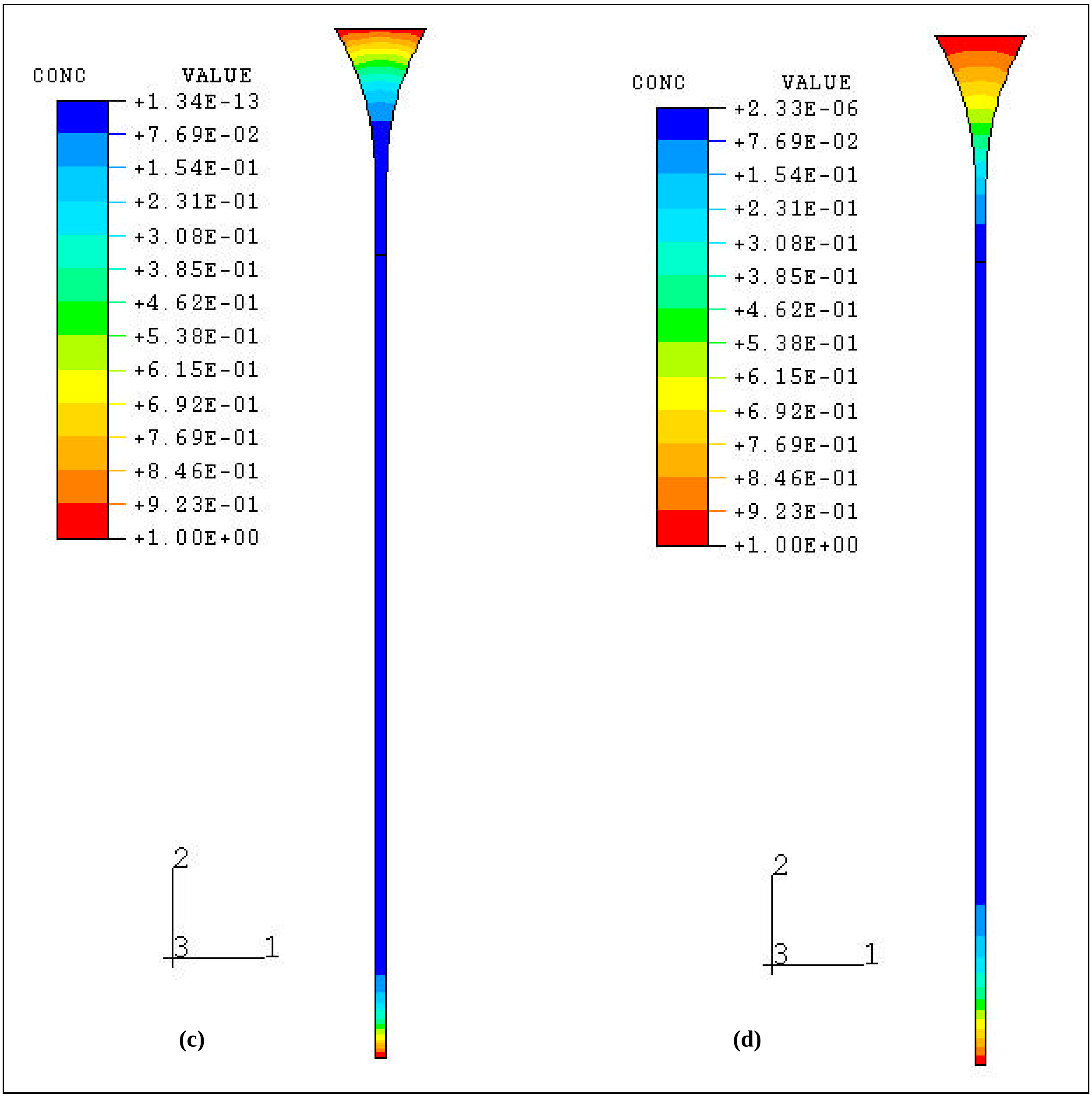


Figure A.2 *- Contour plot of normalised moisture concentration for T-peel joint after:*

(c) 20.4 hours and (d) 79.7 hours

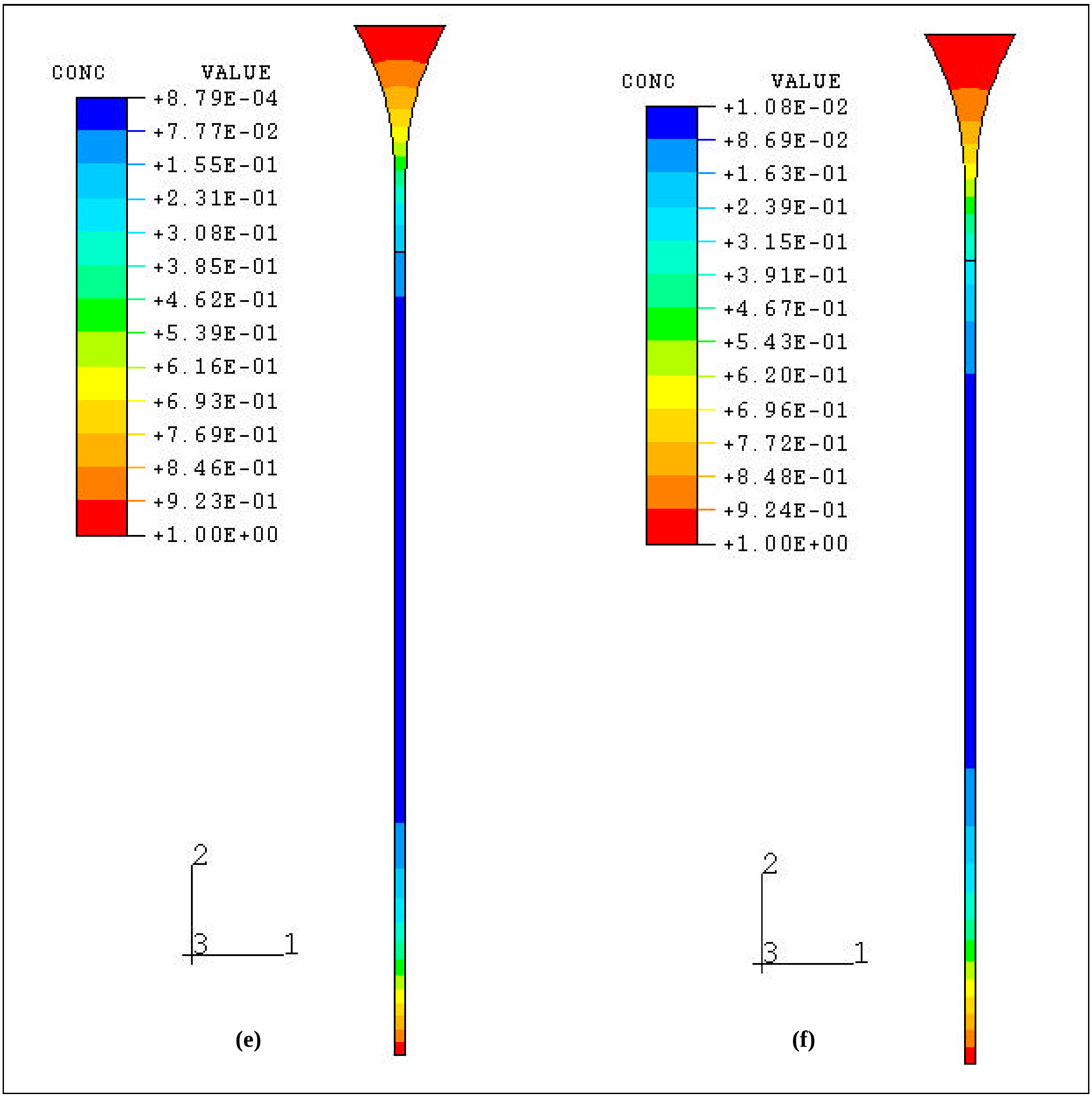


Figure A.3 - *Contour plot of normalised moisture concentration for T-peel joint after:*

(e) 169 hours and (f) 288 hours

APPENDIX C:

RESULTS FROM TRANSIENT DIFFUSION ANALYSIS OF T-PEEL JOINT (100% FILLET)

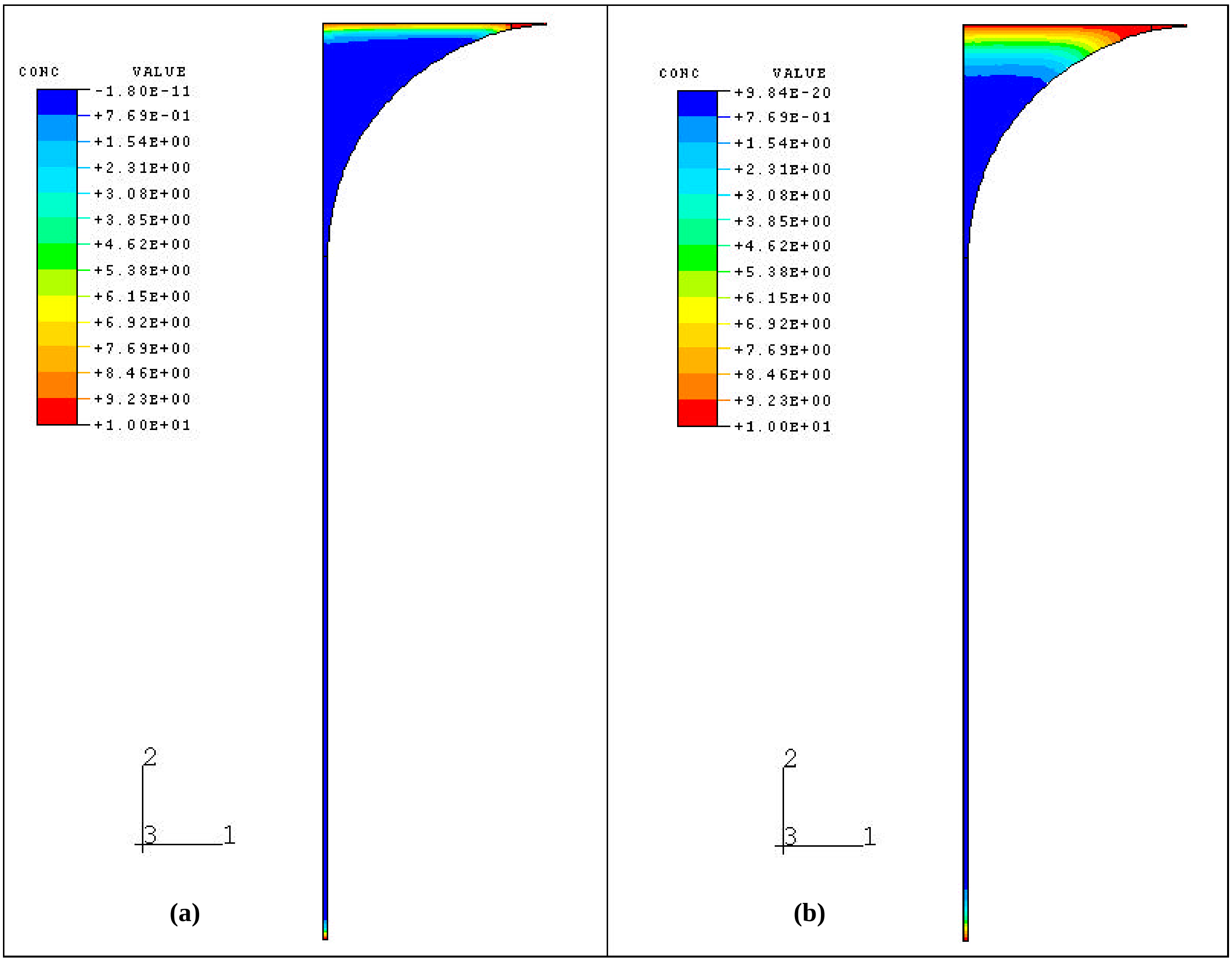


Figure C.1 *- Contour plot of normalised moisture concentration for T-peel joint after:*

(a) 1.05 hours and (b) 12.6 hours

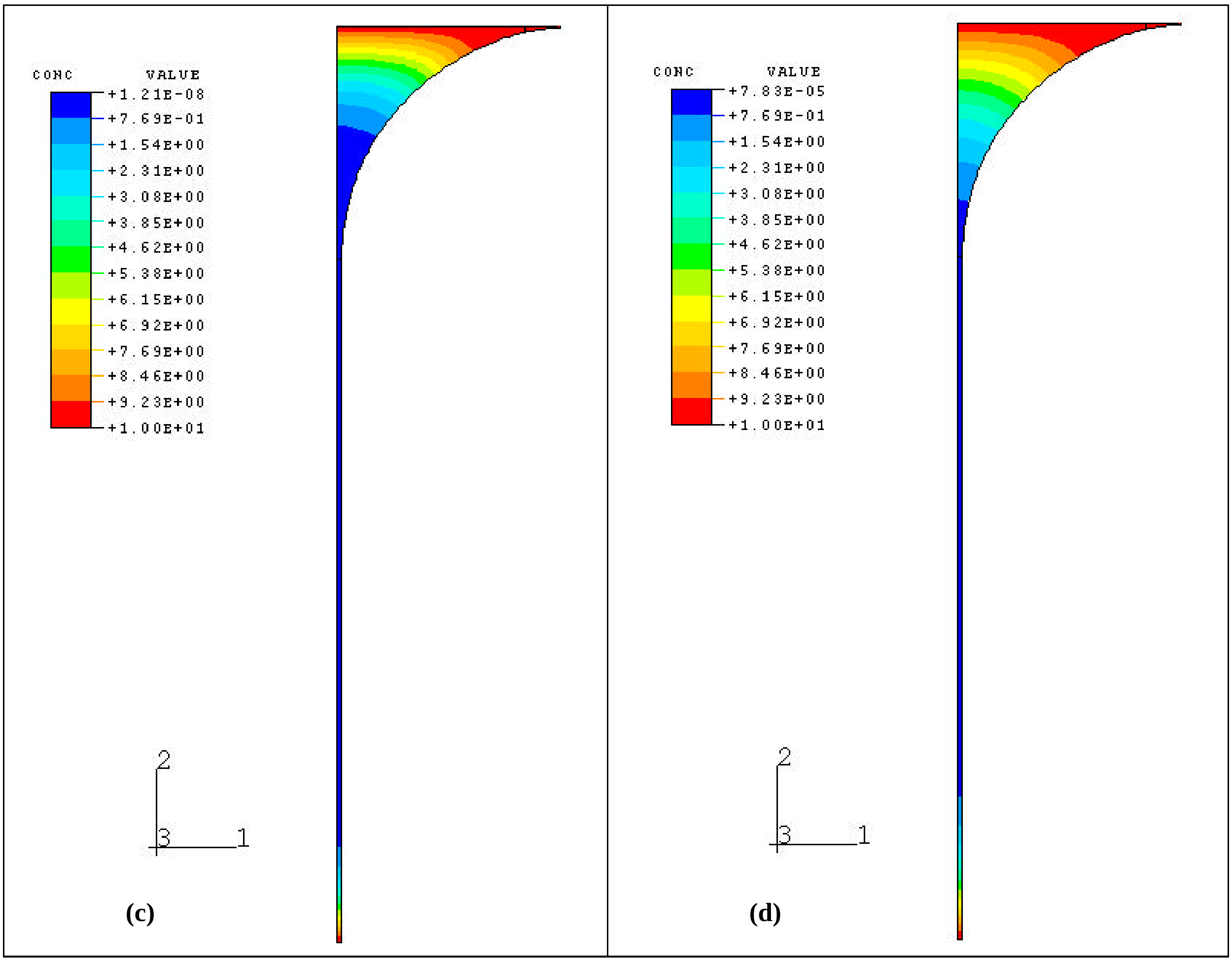


Figure C.2 *- Contour plot of normalised moisture concentration for T-peel joint after:*

(c) 46.1 hours and (d) 105 hours

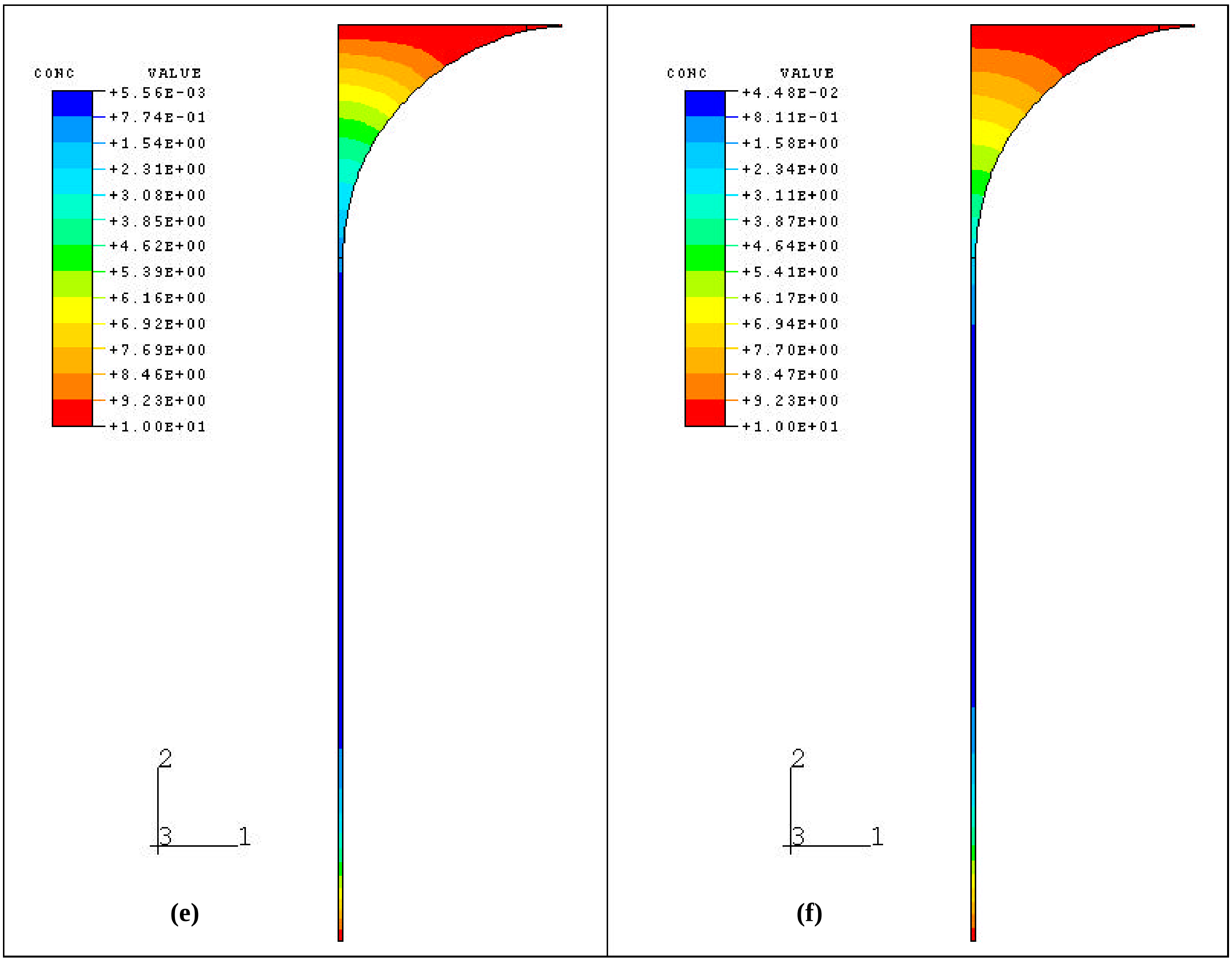


Figure C.3 **-** *Contour plot of normalised moisture concentration for T-peel joint after:*

(e) 189 hours and (f) 288 hours

APPENDIX D:

RESULTS FROM TRANSIENT DIFFUSION ANALYSIS OF PERFORATED SINGLE-LAP JOINT

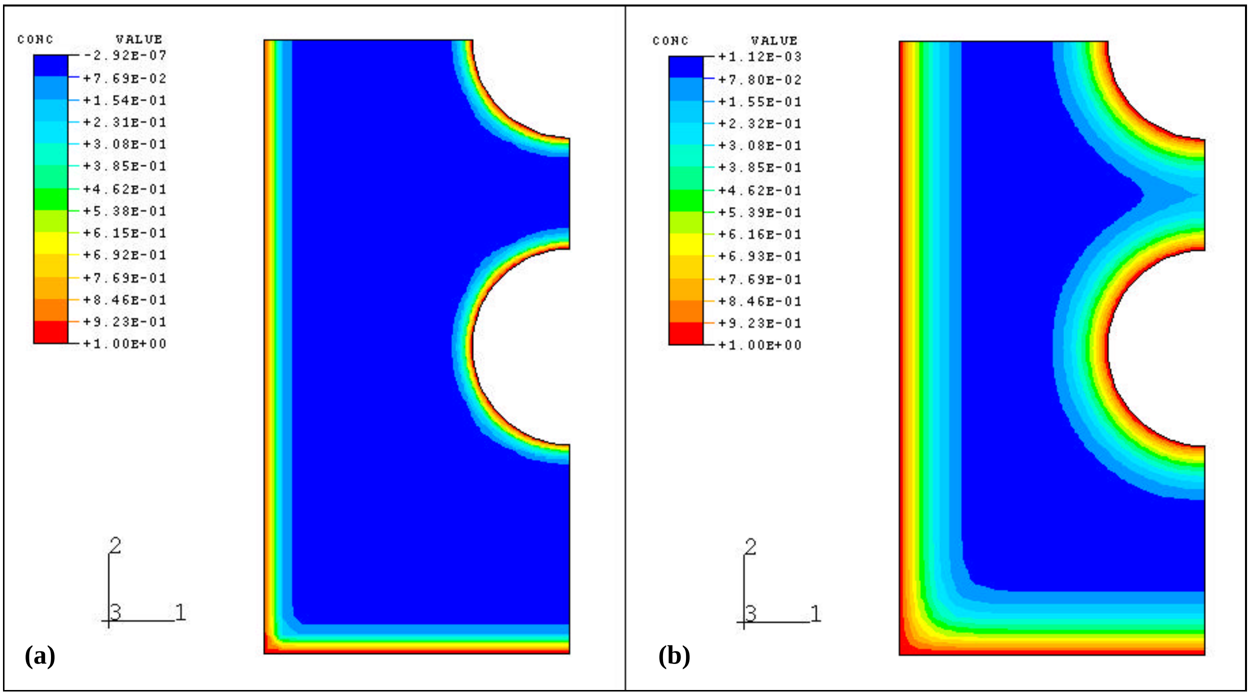


Figure D.1 - *Contour plot of normalised moisture concentration for perforated single-lap joint after:*

(a) 1.0 hours and (b) 9.6 hours

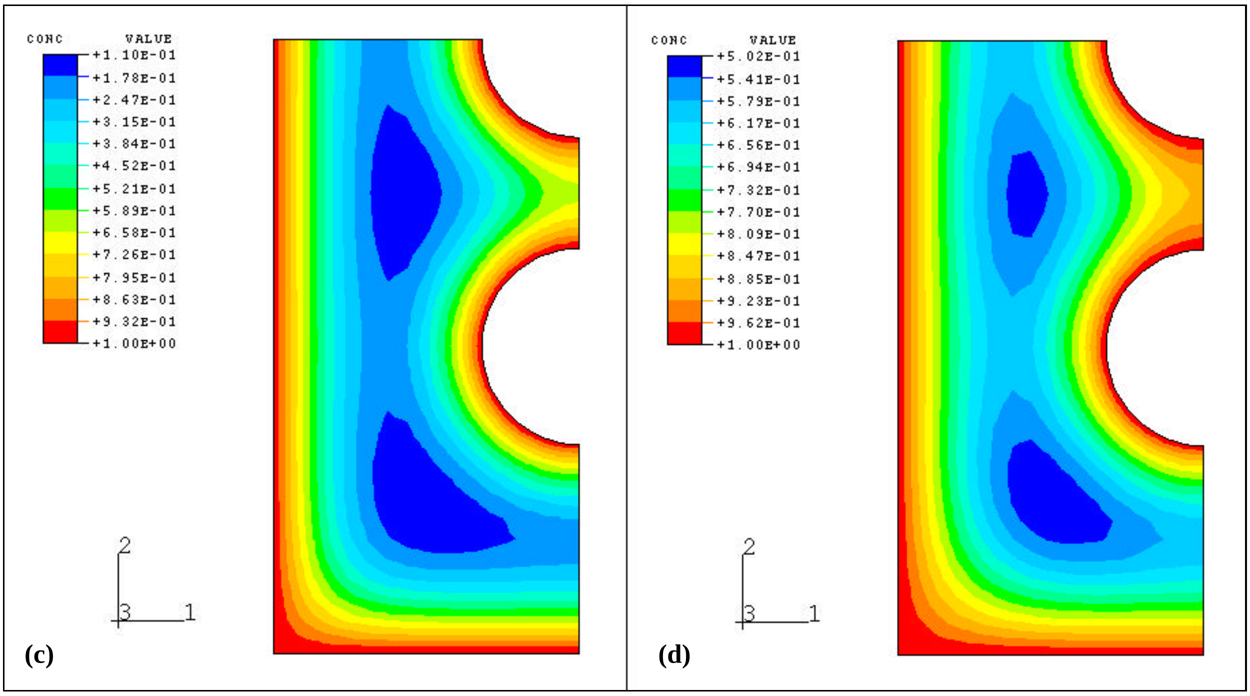


Figure D.2 - *Contour plot of normalised moisture concentration for perforated single-lap joint after:*

(c) 40 hours and (d) 110 hours

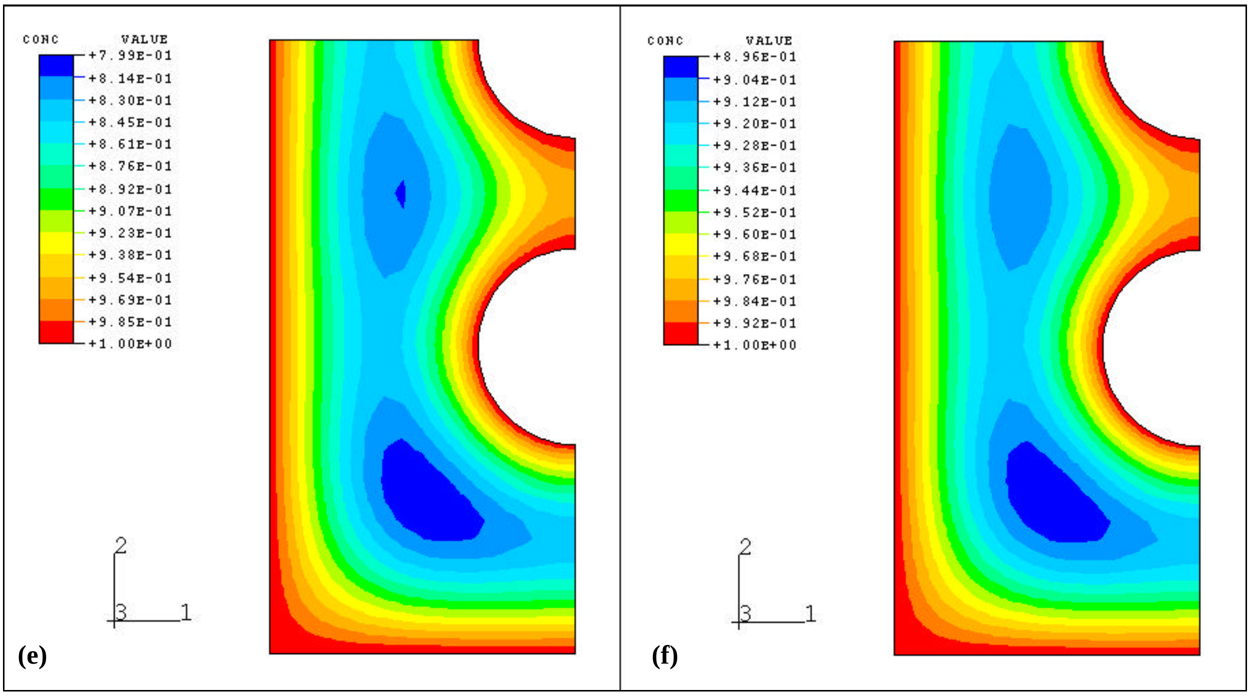


Figure D.3 *- Contour plot of normalised moisture concentration for perforated single-lap joint after:*

(e) 213 hours and (f) 288 hours