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Purpose and Problem Statement

The Lead-Bismuth eutectic (LBE) has been determined from previous experimental studies by the Russians and the European scientific community to be a potential material that can be used as a spallation target and coolant for the TRP proposed application. Properly controlling the oxygen content in LBE can drastically reduce the LBE corrosion to structural steels. However, existing knowledge of material corrosion performance was obtained from point-wise testing with only very sparse experimental data. Scientists have noticed that the concentration of oxygen dissolved in the liquid alloy could control the corrosion rate of steels exposed to Pb or Pb-Bi. At high oxygen concentration, an oxide layer could be formed on the steel surface (lead oxides are less stable than iron oxide), which protects it from corrosion. At low oxygen concentration, there is no oxidation and corrosion occurs by dissolution of the steel components in the liquid metal. The surface of the oxide layer in contact with the bulk flow of liquid metal may also be eroded under a high fluid velocity. Then the surface of the metal will no longer be protected because a porous oxide layer will be formed.

The first subtask of this project involves using a CFD code (3-D simulation) such as STAR-CD to obtain averaged values of stream wise velocity, temperature, oxygen and corrosion product concentrations at a location deemed close to the walls of the LBE loop at more than one axial location along it. The oxygen and corrosion product inside the test loop will be simulated to participate in chemical reactions with the eutectic fluid as it diffuses through towards the walls. Details of the geometry of these loops will be obtained from scientists at LANL. These values will act as a set of starting boundary conditions to the second task.

The second subtask and the more important objective of this project is to use the information supplied by the first task as boundary conditions for the kinetic modeling of the corrosion process at the internal walls of the test loop. The outcome of the modeling will be fed back to the first subtask, and the steady state corrosion/precipitation in an oxygen controlled LBE system will be investigated through iterations. The information is hoped to shed some light on the likely locations for corrosion and precipitation along the axial length of parts of the test loop.
Personnel

Principle Investigator:
• Dr. Samir Moujaes (Mechanical Engineering)

Co-Principle Investigator:
• Dr. Yitung Chen (Mechanical Engineering)

Students:
• Mr. Narain Armbya, M.S. Graduate Student, (Mechanical Engineering)
• Mr. Guanjun Li, Ph.D. Graduate Student, (Mechanical Engineering)

National Laboratory Collaborator:
• Dr. Ning Li, Project Leader, Lead-Bismuth Material Test Loop, LANL
• Dr. Jinsuo Zhang, Post Doctoral Candidate, LANL

Administrative Issues:

We have met with Dr. Joe Smith (June 7, 2004) from the Adapco Co. (providers of STAR_CD). He offered us some very helpful tips on how to setup our grid for the user supplied subroutine that the group is trying to develop for use in the chemical-kinetics simulation. One of our students Narain was out on vacation from May 15-June 21. Guanjun has been having a medical problem which till now has not been well diagnosed.

Technical progress:

Several CFD runs have been made by Narain to simulate flow in pipe fittings. These include sudden contraction, sudden expansion and a T-joint. Testing of a new turbulent model is also being made namely the k-e Chen model which will work a little better with high Re number flows and will be able to predict some of the peculiar flow features relevant to sudden expansions where eventually vortex generation is expected at the backward step (i.e. sudden expansion location) in that flow. It is important to try to predict that because it may have a bearing on the behavior of the chemical kinetics model when it is completed.

Guanjun continues to develop this user subroutine that will allow us eventually to simulate the corrosion/precipitation processes and predict their maximum/minimum location in a typical LBE loop.

Introduction:

Liquid lead-bismuth eutectic is considered as a prototype target and coolant for the Transmutation Research Project (TRP). It is an alloy of 45% lead and 55% bismuth with the melting temperature of 123.5°C and boiling temperature of 1670°C. Using liquid lead-bismuth eutectic (LBE) as coolant in nuclear systems has been studied for more than 50 years. LBE has many unique nuclear, thermo physical and chemical attributes that are attractive for nuclear coolant applications. This liquid’s relatively low melting point and high boiling point in addition to good heat transfer properties make it a very good
candidate for coolant. In addition, lead and bismuth can produce copious spallation neutrons when bombarded with energetic protons. This makes LBE one of the top candidates for a high-power spallation target in an Accelerator-driven Transmutation of Waste (ATW) system. Besides, the use of heavy liquid metal like LBE as a coolant for fast reactors offers several safety and economic advantages. These arise from the following basic material characteristics: chemical inertness with air and water, high atomic number, high boiling temperature and low vapor pressure at operating temperatures. Specifically, heavy-metal coolants do not react energetically with air and water; therefore, coolant fires are not possible and an intermediate heat transport loop is unnecessary. Also, the hard neutron spectrum achievable with these coolants enables the design of cores with minimal neutronic reactivity swing, small control requirements and long neutronic life time. The significantly lower reactivity associated with hypothetical voiding of the coolant, as compared to sodium, makes it possible to design lead or lead-bismuth-cooled cores with a negative coolant void coefficient, thereby eliminating the possibility of severe accidents from consideration. Finally, lead or lead-bismuth coolants provide better shielding against gamma-rays and energetic neutrons, so that less shielding structures are needed. Liquid spallation source also eliminates some of the structural damage problems associated with the targets. Combining the target and coolant roles in one material allows for a simple target design.

One of the critical obstacles to the wide use of LBE as a nuclear coolant, though, is corrosion. The corrosion processes need to be controlled and reduced or they lead to severe safety problems. Unprotected steel undergoes severe attack by liquid lead and lead-bismuth alloy by dissolution of its components in the liquid metal. During the last years, not much was known about possibilities to improve the compatibility of steel with liquid Pb and Pb/Bi. Some compatibility tests with ferritic steels were reported which revealed corrosion attack can be minimized if an oxide layer exists on the steel surface. Scientists at IPPE, Obninsk, Russia, discovered that if an oxide film is allowed to form on the steel surface it prevents corrosion. This protective film consists mostly of steel components’ oxides and it is based on Fe₃O₄. Formation and longevity of this protective film depends on oxygen concentration on the liquid metal. In order to use liquid lead-bismuth in AAA facility, we need to know how to control corrosion of structural materials.
The active oxygen control technique exploits the fact that lead and bismuth are chemically less active than the major components of steels, such as Fe, Ni, and Cr. By carefully controlling the oxygen concentration in LBE, it is possible to maintain an iron and chrome oxide based film on the surfaces of structural steels, while keeping lead and bismuth from excessive oxidation that can lead to precipitation contamination. The oxide film, especially the compact portion rich in Cr, effectively separates the substrates from LBE. Once this oxide film is formed on the structure surface, the direct dissolution of structural materials becomes negligible because the diffusion rates of the alloying components are very small in the oxides. In this circumstance, the only effective means of transferring structural materials into LBE is through the reduction of the oxide film at the interface of the film and LBE. The Los Alamos National Laboratory’s Accelerator-driven Transmutation of Waste (ATW) applications and the Department of Energy’s TRP program have invested in developing LBE technology from spallation target and nuclear coolant applications since 1997. A Materials Test Loop (MTL) has been set up in Los Alamos. The MTL is a facility designed to test the safe operation of a medium-size, forced circulation LBE system with representative thermal hydraulic conditions (as spallation target and/or transmutation blanket systems), to perform corrosion tests, and to develop candidate materials with oxygen control (and related probes and control systems). Figure-1 shows the skeleton representation of the MTL.

It has been well known that fluid flow influences corrosion in many ways, including the increase of the diffusion of reactant species and the transport of potentially protective corrosion product forming ions away from surface. In the mass transfer
controlled regime, the corrosion rate is determined by the mass transfer coefficient and the gradient between the corrosion product concentration at the solid-liquid interface and the concentration in the bulk flow. Corrosion rate is typically a function of local temperature and flow velocity. However, corrosion and precipitation rates and distributions can depend strongly on the global temperature distribution, limiting the applicability of many corrosion models.

The present study involves the estimation of corrosion in the liquid metal, by imposing an analytically developed concentration expression on the wall surfaces and thus benchmarking the CFD tool and performing a series of parametric studies on the loop model. The concentration and temperature diffusions due to different flow regimes have been studied. Regions of maximal corrosion and precipitation have been deduced from the simulations and the results have been compared with the analytical models. STAR-CD has been chosen as the CFD code for this purpose.

**Numerical Simulation Technique:**

The STAR-CD computer simulation code was chosen for the purpose of performing the Computational Fluid Dynamics (CFD) calculations for this project. STAR-CD is a commercially available code that is offered by ADAPCO Co. out of New York State. The code is a transient multidimensional simulator for Thermal hydraulics and chemical reactions occurring in the fluid flow itself.

STAR-CD is a general-purpose code that solves numerically a set of differential equations that describe the following conservation laws: mass conservation, momentum, energy and chemical species. The following equations are solved by this code:

**Continuity Equation:**

\[ u_{i,i} = 0 \]  

**Momentum Equation:**

\[ \rho_0 \left[ \frac{\partial u_i}{\partial t} + u_j u_{i,j} \right] = -P_j + \mu \left( u_{i,j} + u_{j,i} \right) \]  

**Energy Equation:**

\[ \rho C_p \left( \frac{\partial T}{\partial t} + u_i T_j \right) = (K \ast T_j) \ast + \mu \Phi \]  

**Species Transport:**

\[ \rho \left( \frac{\partial C}{\partial t} + u_i C_{n,j} \right) = \left( \rho \alpha_n C_{n,i} + q_{c,i} + R_n \right) \]  

Due to the Re number estimate for flow in a LBE loop a turbulent flow model should be used as a constitutive model for the momentum transport. It was decided that a k-\( \varepsilon \) model is to be used to account for that behavior. The model consists of adding two more non-linear (transport equations) partial differential equations to each unknown nodal location. The \( k \) denoted the turbulent kinetic energy \( u_i u_j \) and the \( e \) is the viscous dissipation rate of the turbulent kinetic energy \( u_{i,j} u_{i,j} \). The resulting equations are:
k – transport equation:
\[ \rho_o \left( \frac{\partial k}{\partial t} + u_i \frac{\partial k}{\partial x_i} \right) = \left( \mu_o + \frac{\mu_t}{\sigma_k} \right) \frac{\partial^2 k}{\partial x_j \partial x_j} + \mu_t \Phi + \frac{\beta_t}{\sigma_t} \frac{\partial T}{\partial x_j} - \rho_o \varepsilon \]  \hspace{1cm} (5)

\varepsilon – transport equation:
\[ \rho_o \left( \frac{\partial \varepsilon}{\partial t} + u_j \frac{\partial \varepsilon}{\partial x_j} \right) = \left( \mu_o + \frac{\mu_t}{\sigma_k} \right) \frac{\partial^2 \varepsilon}{\partial x_j \partial x_j} + \frac{c_t}{k} \mu_t \Phi + \frac{c_t(1 - c_t)}{k} g_i - \rho_o c_z \frac{\varepsilon^2}{k} \]  \hspace{1cm} (6)

**Benchmark Study:**

Benchmark is important in research, especially in numerical simulation. It provides the validation of the tools and the base for the further effort. Before used to carry out calculation for more complicated cases, the code was applied to a classic problem and compare outcome with widely accepted results. Incompressible flow in sudden expansions is one of the classical problems and suits our calculation domain perfectly. The other fittings considered are the t-joint, the sudden contraction and the elbow.

The first section sheds light on the concentration and temperature profiles obtained from the sudden expansion model. Sudden contraction model is dealt in the second section. The third section sheds light on the concentration and temperature profiles obtained from the t-joint model. The fourth section deals with the elbow. The results are shown for the flows in the turbulent regime of Re=200,000.

**Sudden Expansion**

A model of sudden expansion is created. The diameter at the inlet was selected as 0.0254m. The lengths of the inlet and outlet regions are taken as 10 diameters. The ratio of the inlet to outlet diameter is 1:2. The aspect ratio varies between 8 and 10 as specified by the CFD package. Runs were simulated for the Reynolds number of 200,000. The simulated results obtained are as shown. The imposed wall temperature varies all through out the length of the MTL. For this study, the temperature along the fitting length is taken as 723K. The fluid enters the inlet at 623K. Figure 2 shows the variation of the temperature profile along the fitting length.
The diffusion of the temperature into the fluid is clearly visualized in the above figure. Immediately after the sudden expansion, the diffusion of the temperature is prominent in the transverse direction and as the flow develops, diffusion is prominent in the transverse direction as is expected at higher Re numbers.

Figure 3 shows the concentration profile of LBE in the fitting. The diffusivity of iron into LBE is as low as $10^{-8}\text{m}^2/\text{s}$, which makes the diffusion very slow. Figure 4 shows the graph of concentration gradient v/s distance from the inlet. These gradients represent corrosion/precipitation on these locations. The decrease in the concentration gradient is due to the flow reversal, which takes place after the sudden expansion followed by an increase and then a decrease at further distances downstream. This is due to the formation of new boundary layers after the sudden expansion region.
Figures 4 and 5 show that the change in concentration is proportional to the change in temperature.

The next important result is the establishment of grid independency. Here the layer of cells next to the wall in the sudden expansion model were isolated and refined. This layer was first divided into 5 more layers and later on into 10 and 15 layers. The result obtained is as shown in Figure 6. It shows that the grid independency has been achieved.
Figure 6. Results of grid independency test for sudden expansion model.
Concentration gradient on the wall

T-joint Model

The inlet and the outlets’ diameter is 0.0254m. The length of the main arm is 0.0762m. The length of the branch of the t-joint is 0.254m. Figure 2 shows the t-joint model.

For this study, the temperature along the fitting length is taken as 723K. The fluid enters the inlet at 623K. The outflow percentage for each of the outlet arms is taken as 50%. There are two cases for this model. One is when the inlet is from the straight arm and the other when the inlet is from the branch arm. Results of both the cases have been presented.

First the results obtained from the inlet from straight arm are presented.
The fluid enters the inlet at 623K. The wall temperature is maintained at 723K. Figure 8 shows the concentration profile. The wall concentration is a function of wall temperature. The diffusion is more prominent in the flow reversal region in the branch of the t-joint. Figure 9 shows the graph of concentration gradient along the straight wall. Point A in the graph is the area where the branching starts. In this region there is an increase in the concentration gradient because of the regeneration of the boundary layers after the branching.
Point E in Figure 10 is the elbow of the T-joint. There is a decrease in the concentration gradient because of the flow reversal. F here is the location where the inlet flow comes in the T-joint.

Now the results for inlet from the branched arm are presented. Figure 11 shows the flow pattern when the inlet is through the middle arm. It is noted that there is a stagnation region as expected at the region near point A of figure 7. The velocity vectors right outside that region show a larger magnitude and connect to the outer wall of the tee intersection on both sides. Two reversal flow zones are noticed on the inner wall of the T-joint right downstream of the right-angled intersection of the two T-sections. The symmetry of the flow is also observed as expected. The flow starts to redevelop downstream but the simulation problem solution field in not long enough to capture that.
Figure 12 shows the concentration profile for this fitting. The diffusion of temperature and concentration is more prominent away from the flow reversal regions. The diffusion of concentration does not seem to be affected by the stagnation zone.

![Concentration profile for T-joint at Re = 200,000](image1.png)

The graph in Figure 13 showing variation of the concentration gradient on the wall is plotted along the line AYB as shown in Figure 7. When the fluid enters through the middle arm and impinges on the opposite wall an area of flow stagnation is created. Point A is the area of flow stagnation. A decrease in the concentration gradient occurs as the fluid comes out of this region and as the new boundary layers are formed, the concentration gradient increases. But as the flow progresses, it encounters the region of flow reversal which results in the decrease of concentration gradient. Point Y indicates the flow reversal region.

![Concentration gradient v/s Distance From Center to bottom exit of T-joint fitting](image2.png)

Figure 13. Concentration gradient v/s distance from center to the bottom exit.
The graph in Figure 14 is plotted along the line DEF (elbow). The region EF is where flow reversal occurs. Hence a marked decrease in the concentration gradient occurs from what essentially is initially a fairly stable value of the gradient as the flow is coming through a short straight tube. A slight increase is noticed as the flow straightens out after E, which is indicative of the boundary layer reattaching on the wall.

**Sudden Contraction**

The model, which had been created for the sudden expansion, is used here. But the inlet and outlet are changed accordingly. The simulated results obtained the turbulent flow is as shown. Figure 15 shows the temperature profile for the sudden contraction fitting and figure 16 shows the temperature gradient along the length of the fitting. The temperature and the concentration are maintained as that of the sudden expansion model. The results shown are for Re=200,000.
Figure 15. Temperature profile for sudden contraction.

Figure 16. Temperature Gradient along wall v/s Distance from Inlet

The temperature diffusion into the fluid can clearly be observed in the Figure 15. The sudden jump observed in the graph is the region of sudden contraction. Figure 17 shows the graph of concentration gradient v/s the distance from inlet for the sudden contraction model. This is similar to the graph in Figure 16. The concentration gradient decreases as the fluid enters the fitting and as the fluid encounters the sudden contraction region the concentration gradient increases as new boundary layers are being formed and then it evens out.
The following graph shows the result of grid independency test. The test was carried out in the same way as that of sudden expansion. It shows that the grid independency has been achieved.

Figure 16. Concentration Gradient along wall v/s Distance from Inlet

Figure 17. Results of grid independency test for sudden expansion model. Concentration gradient at wall
Study of Surface Chemical Reactions

The original straight pipe (0.05m * 2.0 m) 3-d model has been modified. Specifically, the CFD domain, material, fluid zones, each species’ inlet concentration, etc., were kept the same while the boundary conditions of 2 sides of the baffle cells are changed to constant heat flux boundary conditions. In addition, the temperature of interface between outer fluid zone (green area shown in Figure 18) and wall was changed to 1400 k. This liquidized the iron adjacent to the inner surface. This way, the homogeneous chemical reaction between liquidized Fe and O2 took place and was used to be an approximate substitute for surface chemical reaction. Figure 19 shows the updated model.

The chemical reaction subroutine dealing with surface chemistry has been updated. The updated subroutine is shown in Appendix (latest version of subroutine updated in August, 2004). Arrhenius Equation was still used to calculate the reactant reaction rate but some constants and coefficients used in calculating chemical reaction rate have been updated using some empirical data found in some chemical reaction research paper and text books. The species' molecular weights, the initial species’ concentration, iterations, etc., did not change.

Several run with and without subroutine connected have been made. Figure 20 shows velocity profile inside the piping without subroutine connected. It is obvious that this is a laminar flow. Figure 21 shows velocity profile inside the piping with subroutine connected. It is interesting to see that the flow becomes more regular.

Figure 22 shows Fe3O4 concentration distribution profile across the pipe when subroutine is activated. It is observed that there is a concentration gradient in radial direction inside the pipe. The area with the highest concentration occurs in the baffle cell which is adjacent to iron surface while the area with lowest concentration occurs around the central line of the pipe. It can be inferred that after Fe3O4 is formed, it diffuses to the fluid area close to the central line of pipe. But, overall, the concentration value is very low.

Figure 23 shows LBE liquid temperature distribution inside the pipe when subroutine is activated. It is also observed that there is a temperature gradient in radial direction inside the pipe. Similar to concentration gradient, the area with the highest temperature occurs in the baffle cell which is adjacent to iron surface while the area with lowest temperature occurs around the central line of the pipe. It is obvious, based on the model, that heat was transferred from inner wall to central area of the pipe due to the setting of constant heat flux boundary conditions of baffle cells. Since the temperature is not high, it could be inferred that the chemical reaction did not generate too much heat.

Briefly, the chemical reaction model subroutine worked well and this simple CFD model roughly simulated the chemical reaction, diffusion, and heat transfer condition inside the LBE piping. Since the subroutine works well, it can be used in much more complex model.
Figure 18 The original 3-D CFD model (0.0508 m * 0.25 m straight pipe)

Figure 19 The updated 3-D CFD model
Figure 20 The velocity vector inside the pipe when subroutine is not linked
Figure 21 The velocity vector inside the pipe when subroutine is linked
Figure 22 Fe3O4 Concentration distribution contour across the pipe
Figure 23 Liquid temperature distribution contour across the pipe
Appendix (latest version of subroutine updated in August, 2004)

LBE surface chemical reaction subroutine
C**********************************************************************
SUBROUTINE REACFN(RATE)
C     CHEMICAL REACTION RATE
C**********************************************************************
C---------------------------------------------------------------------*
C     STAR RELEASE 3.150
C---------------------------------------------------------------------*
INCLUDE 'comdb.inc'
COMMON/USR001/INTFLG(100)
INCLUDE 'usrdat.inc'
COMMON/USREAL/IR,NR
COMMON/USREAR/TAUL,TAUG,AEBM,BEBM,AMFU,AMFB,ARCK,BETCK,EA
CT, RTCKF,AMRC(3),RTCKR(3)
common /speed03/ wmfuu,wmoxi,stoxi,tauml
DIMENSION SCALAR(50), HFORM(50)
EQUIVALENCE( UDAT12(001), ICTID )
EQUIVALENCE( UDAT03(001), CON )
EQUIVALENCE( UDAT04(001), CP )
EQUIVALENCE( UDAT04(002), DEN )
EQUIVALENCE( UDAT04(003), ED )
EQUIVALENCE( UDAT04(006), P )
EQUIVALENCE( UDAT04(008), TE )
EQUIVALENCE( UDAT04(009), SCALAR(01) )
EQUIVALENCE( UDAT04(059), U )
EQUIVALENCE( UDAT04(060), V )
EQUIVALENCE( UDAT04(061), W )
EQUIVALENCE( UDAT04(062), VISM )
EQUIVALENCE( UDAT04(063), VIST )
EQUIVALENCE( UDAT04(007), T )
EQUIVALENCE( UDAT04(067), X )
EQUIVALENCE( UDAT04(068), Y )
EQUIVALENCE( UDAT04(069), Z )
EQUIVALENCE( UDAT09(001), IS )
EQUIVALENCE( UDAT10(101), HFORM(01) )
C---------------------------------------------------------------------*
C For a specified material “imat” (i.e., surface cells where
C reaction takes place) and for a specific reaction “IR”, set the
C necessary parameters (i.e., initial concentrations, temperature, etc.)
C----------------------------------------------------------------------
    if(imat.eq.1) then
      If(IR.EQ.1) then
        iFe3O4=1
        iPbO=2
        iO2=3
        iFe=4
        iPb=5
        wFe3O4=232
        wPbo=223
        wO2=32
        wFe=56
        wPb=207
        sum=0
    C----------------------------------------------------------------------
    C  collect all the scalars for the reaction rate
    C----------------------------------------------------------------------
    Sum  = scalar(Fe3O4)/wFe3O4 + scalar(PbO)/wPbO +
       &   scalar(O2)/wO2
       &   + scalar(Fe)/wFe + scalar(Pb)/wPb

    wmix=1./sum

    T1=T
    s1=scalar(iFe3O4)
    if(s1.lt.0) s1=0
    C----------------------------------------------------------------------
    C  Some constants in Arrhenious expressions were updated in LBE case
    C----------------------------------------------------------------------
    rate=1.84e7*exp(-25000./(1.987*T1))
    rate=rate*(den*scalar(iFe3o4)*wmix/wFe3O4)**.75
    endif
    endif
    Return
    End
References