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Review Article

Computer simulation tools as applied to radioisotope production target design

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Summary

Computer simulations have been applied to many isotope target design problems with mixed results. At best, computer simulations provide rapid evaluation of design alternatives with high accuracy and without the cost in time and material of 'cut-and-try' methods. However, models are only helpful when well posed, well validated, and theoretically sound. In this context, the growing availability of commercial simulation packages provides an opportunity for the development of more advanced target systems. Copyright © 2002 John Wiley & Sons, Ltd.

Key Words: isotope targets; modeling; simulation

1. Introduction

The engineering of targets for the accelerator production of radionuclides is a multidisciplinary effort. In order to be successful the designer must take into account the ionization heating of the target materials including heat transport through the target medium, through supporting equipment (windows, target bodies, collimators) and into coolant. In addition, the radiation chemistry in the target material, fate of recoils of the desired and undesired products, monitoring of deposited charge including suppression of or correction for secondary

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currents, and shielding of resultant radiation flux must all be considered. The resulting constraints on materials, be they yield strength, ductility, chemical compatibility, corrosion resistance, or melting point, drive the final design.

Because of the complex nature of the problem, some computer modeling can be useful. Of course where calculations by hand will do, they should be used in favor of computer models due to the expense of codes and time taken to prepare data sets. However, where the geometry of the problem is complex (e.g. heat transport in a 3-D target support structure), computer simulation is appropriate. Where there are multiple simultaneous constraining equations (e.g. 2-phase flow, chemical reaction dynamics), simulations are also useful. However, they can be misleading if the models, boundary conditions or simplifying assumptions are incorrect or inappropriate. Therefore, computer models should be validated for a case similar to the one in question, and then used to explore relative changes in geometry, material, or operating conditions. Only in the best circumstances do computer models render absolutely accurate information.

Various computer simulation tools available or developed for isotope targets can be grouped into three broad categories. The most widely modeled process in target operation is the transport of heat in the targets and their enclosures. Some of these models have included mass transport (flow) on a macroscopic level. Models have also been constructed to simulate the transport of mass on a microscopic level in terms of particle motion (stopping and recoils). In target, chemistry has also been modeled using computational methods.

2. Heat transport

The modeling of heat transport in isotope production targets may be as simple as a multi-compartment, non-dimensional custom model, through finite element simulations requiring significant computational resources. In the latter case, a two or three-dimensional physical model is segmented into multiple elements, and the specified differential equations are linearized and solved for each geometrical element. In the simplest case, heat transport through one or several solids is modeled by solution of the Fourier heat conduction equation:

$$q = -k\nabla T \tag{1}$$

In anticipation of the upgrade for the Brookhaven Linac Isotope Producer (BLIP), Mausner *et al.*[1] used ANSYS to model heat flow through three target materials; zinc powder (production of ^{67}Cu), RbCl pellets (production of ^{82}Sr) and liquid gallium (production of ^{68}Ge). The simulation was in two dimensions (cylindrical or r, z coordinates) and did not take phase change or flow into account. External heat transport (into the coolant) was not modeled, but reasonable boundary conditions were set based on general engineering practice. The finite element models were useful in several regards. For the liquid target they predicted temperatures well below boiling. For the powder targets they predicted central target temperatures above boiling, which drove further simulation and design changes.

In subsequent work, Mausner and Hock[2] used the ANSYS code to evaluate design alternatives including different target materials, thinner target enclosures, embedded conductive vanes, off-normal angle of incidence and beam shaping or wobbling. In the end, a thinner target enclosure with different target material and flatter beam shape were selected. No flow modeling was reported for the BLIP targets.

Nortier *et al.*[3] carried out a thorough and detailed validation of computer thermal analysis of solid targets. The analytical component of the work (performed using ALGOR) included sensitivity studies versus beam shape in two dimensions, and changes in the target rear cooling fin structure. No fluid dynamics modeling was used, but variation of the target face temperature was modeled with coolant flow by adjusting the heat transfer coefficient.

The computational models were validated by the construction of a test target that had both embedded and surface mounted thermocouples. The surface mounted thermocouples were fashioned from a nickel/silver sandwich by selectively nickel plating areas on the silver target back. Data from these thermocouples correlated very well with the FEA models over a range of beam currents. The authors projected a doubling of the current they were using routinely from the design changes the modeling and experiments had suggested.

Beyond the modeling of heat transfer through solids, Lenz and Vincent[4] used a computational fluid dynamics (CFD) 3-dimensional code[5] to model flows and temperatures in a liquid rubidium target. Aside from the application of CFD to isotope targets, there were several novel aspects of this work. The heat and mass transport inside the cylindrical target were modeled, yielding detailed information about the maximum temperature achieved in the target material for a variety of

beam conditions. Buoyancy driven flow visualizations were presented, showing both a two-fold symmetric (across the vertical plane) circumferential flow pattern and an axial flow pattern as well. Beams of various uniform intensities were examined. Transverse beam shape was not investigated. However, increased dE/dx at the rear of the target due to reduced energy was included in the model. Drops in maximum temperature were predicted with more diffuse beam and with a lowering of the beam centering on the target (correlated with similar experiments with water).[6]

The external coolant flow was also modeled for this target. A half model including an impinging water jet was generated, which yielded typical heat transfer coefficients for the system and highlighted the exit window as the area of highest temperature. Perhaps most novel was the modeling of conjugate heat transfer in the system. All three main elements, coolant, enclosure and contents of the target were modeled simultaneously, including flows both internal and external to the target. The study isolated appropriate beam intensities and centering to maintain temperatures below the boiling point of the rubidium. It also demonstrated flow stability over a wide range of operating currents.

The encapsulated isotope production targets at TRIUMF have been modeled in a similar fashion.[7] Here, again the external and internal flows were combined in a conjugate model. However, the model was 2-D axisymmetric and did not include buoyancy driven flows. No axial dependence on stopping power was modeled. The 3-D fluid module from EMRC, DISPLAY III/NISA II was used.

At the same workshop a much simpler, non-FEA model was presented for heat transfer.[8] This model was built in a spreadsheet program (QuattroPro). Individual spreadsheet cells corresponded to r, z elements, similar to FEA but using a more macroscopic, multi-compartment control volume analysis. Buoyancy driven flow and mass transport inside the target were not modeled directly. However, heat transfer coefficients inside the target were estimated using the Dittus–Boelter formulation for the Nusselt number. The model was adjusted to fit experiment. The result was confirmation of the assumption that helium window cooling did not contribute significantly to the general heat removal mechanisms.

Lenz further applied CFD and other codes to a true two-phase target system for production of ^{18}F from protons on ^{18}O enriched steam.[9] In addition to CFDesign, Lenz used HTPIPE, a steady

state, axisymmetric geometry code developed by Los Alamos National Laboratory to model the vaporization/condensation cycle in this two-phase target.

At CTI, the author has used CFDesign to model the internal heat transport in an enriched water target, as well as the heat transfer to impinging-jet helium cooling on the target windows. In the helium, case results were validated by experiment that show the maximum heat flux is approximately 25 W/cm^2 (Figure 1).

CFDesign is equipped with an enthalpy conservation mechanism that allows modeling of the vaporization of liquid water at elevated pressures in the target. A Gaussian transverse beam profile (70% transmission through an 8 mm circular collimator) was used, along with axial variation of the heat load to include the Bragg peak in the target. The target window was modeled as an insulator based on the earlier results for helium. We did not achieve very good absolute correlation to experiment. In order to obtain the void we expected to see in the target, we had to increase simulated current to much more than 40 μA. However, we did achieve similar results to those obtained by Lenz for the liquid rubidium target, namely, peak target temperatures are reduced by making the beam more diffuse and having the beam maximum lower than the target center. We also observed both flow patterns (axial and symmetric circumferential). Images of the vapor

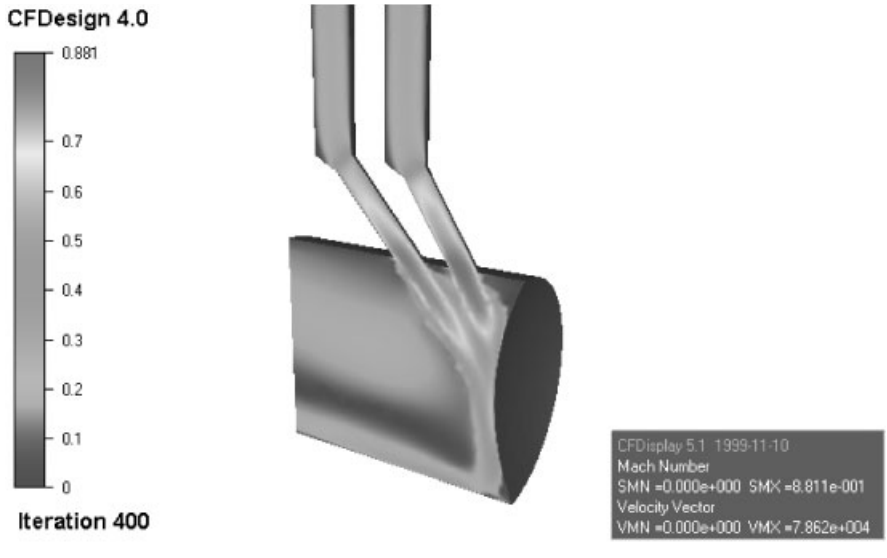

Figure 1. Helium jet simulation using CFDesign

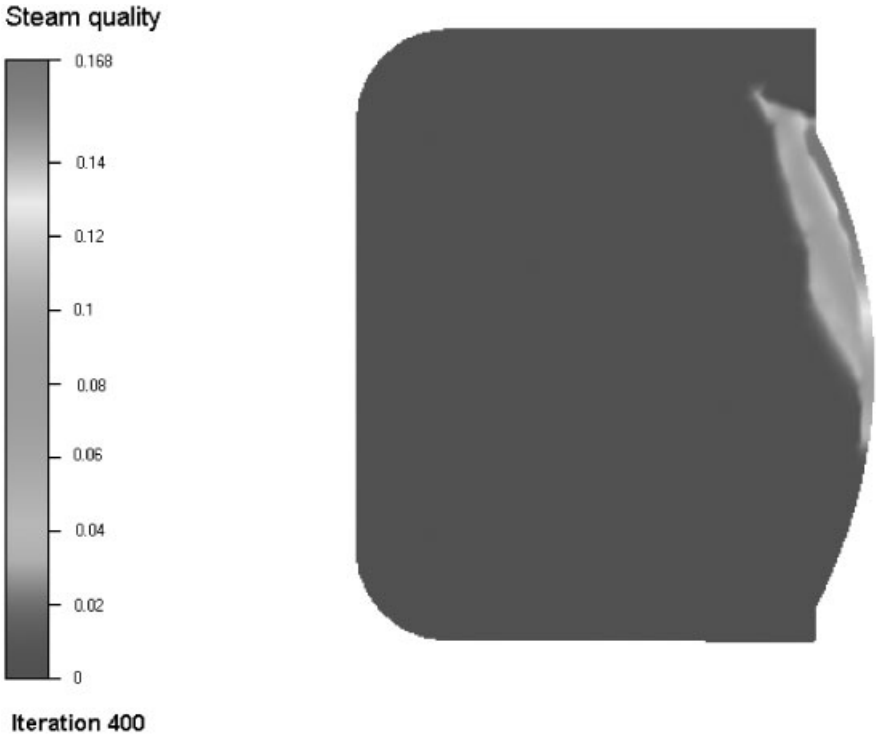

Figure 2. Steam quality in a high-pressure water target

void and its relationship to the target window are confirmed by experience. Figure 2 is an image of the steam quality (percent vapor by mass) in a high-pressure water target. The image is the vertical center plane of the target, with beam entering from the right through the hemispherically deformed target window. The bulk of the target has a steam quality of zero, and is completely liquid. Flashes to steam are only observed near the window and in the upper half of the target. We observe discolorations on the target window very similar to the void image from the computer models.

The near-insulating nature of the target window was confirmed by the simulations and is notable. This surface of the target is under severe thermal stress. Without the target water as coolant, the window temperature increases significantly until radiative heat transfer can accommodate the heat deposited in the window.

Computer models of a large variety of targets with FEA/CFDS codes yield important similarities. The codes are best used to determine relative effects, such as stability of flow patterns over a variety of irradiation conditions, or relative moves of beam maxima in the target.

Absolute results are less likely and should be validated rather than trusted.

3. Particle transport

Codes that simulate mass transport of ions or recoiling nuclei are similarly diverse in complexity and specialization to isotope target simulations. SRIM2000 (Stopping and ranges of ions in matter, formerly known as TRIM) is a group of programs which calculate the stopping and range of ions into matter using a quantum mechanical treatment of ion–atom collisions.[10] The code is available for free distribution (http://www.srim.org), and is an invaluable tool for the calculation of range and radial straggling in isotope targets. It is especially useful in determining the appropriate cone angle for gas targets, and appropriate target thickness if designing a target with some exit energy. The stopping power calculations can be used to estimate range in materials where published data are unavailable. The code assumes a point beam and heating is not modeled.

SUPERP is a more sophisticated multiscattering Monte Carlo code and thermal analysis package.[11] It is designed more specifically for isotope targets based on a stacked foil geometry. Multiple Coulomb scattering is modeled in the MSCT module. Beam transverse dimensions, intensity and emittance and angle of attack are all adjustable. Thermal properties of the assembly are modeled in the HEAT module. Conduction, radiation and convection are all treated.

For a time CTI investigated the utility of two-phase (solid/liquid) target materials based on sol–gel derived carbon foam structures. In order to arrive at a starting specification for pore size and void fraction a simulation algorithm was devised using Microsoft Excel. Reactions were modeled using the compound nucleus model. The simulation was one-dimensional; uniform thickness alternating layers of solid and liquid. Range/energy data for the bombarding particle and the recoiling product nucleus in both solid and liquid came from TRIM. At the entrance and exit of each layer, the fate of recoil products was estimated for forward and backscattered recoils only.

The first application of the algorithm was to proton bombardment of porous carbon pucks and prediction of the recovery fraction of ^{13}N and ^{18}F in aqueous form.[12] The algorithm was suitable for order of magnitude prediction of the recovery of both isotopes. It indicated

correctly that the two geometries selected for the experiment had only moderate differences in their recovery fractions. The same porous carbon materials were later subjected to irradiations with 5 MeV deuterons and 14.8 MeV ^{3}He particles.[13] The model again showed general agreement with measurements in these cases, as well as the proton irradiation of carbon fibers.

W.L. Dunn of Quantum Research Services developed a more sophisticated set of codes, applied to the same two-phase recoil problem. The LAYERTAR (and later FIBTAR) code used TRIM (now SRIM) for energy loss calculations and allowed for input of beam dimensions and transverse intensity.[14] The code assumed multiple plate elements and calculated the recovered activity from recoil escape of ^{15}O from quartz fibers under 27 MeV proton bombardment and subsequent delivery as [^{15}O]ozone. The code predicted values that were larger than measured by a factor of three. There are several reasons for differences in both LAYERTAR and in the spreadsheet model applied to the porous carbon. The geometries are not perfect plates but fibers, spheres and other microcellular geometries. Moreover, chemical form and reactivity of the species in question limit the practical recovery of these isotopes. The effect of in-target chemistry was not a subject of either model.

4. Conclusions

With the advent of extremely fast desktop computation at a reasonable price, tools for computer modeling of any and all physical processes are becoming widespread, and an irresistible aid in the design of complex devices such as isotope production targets. Pitfalls remain in the determination of real and reliable boundary conditions and the simplifying assumptions used to render the problem tractable. Validation of a variety of codes in this arena has been a difficult task and the results are incomplete. Depending on the desired outputs, complex computer codes are beginning to claim small successes. Further work on the refinement and validation of such codes is warranted before they are relied upon for major design decisions, but their utility in limited cases is demonstrated.

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