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CYCLOTRON TARGETRY FOR PRODUCTION OF SHORT-LIVED POSITRON EMITTERS

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Abstract

The basic concepts of cyclotron target design are presented along with the relevant practical experience gained by workers in this field over the years. Results are presented from several recent studies on the temperature and density distribution inside gas and liquid targets.

Introduction

The field of Positron Emission Tomography (PET) has grown enormously in the past 15 years. In 1976, there were only three operating PET centers all of which were in the United States. There are currently more than 60 centers and the distribution is world-wide.

One thing that all these centers have in common is that there must be short-lived positron emitting isotopes at hand and at present this requires that a cyclotron and associated targetry must be somewhere nearby. It is necessary that the targetry systems, whether commercial or built in-house, be reliable and efficient.

The design and building of cyclotron targets is a multi-disciplinary field. It requires the combination of elements from chemistry, physics and engineering. In this report, several aspects of the design of cyclotron targets will be discussed. The sometimes mutually exclusive solutions to the numerous problems encountered in the optimal design of a target will be explored and experience on the compromises made will be shared. There are many more combinations of solutions to the problems than will be given here, so that this is in no way the only way to handle the problems and new and better solutions will be forthcoming in the future. The ways in which the fields of chemistry, physics and engineering relate to the design of targets are described below.

The references given in this work are very general references which are familiar to most workers in the field. It is not the purpose of this paper to review all the contributions which have been made in this field during the past 20 years but only to give an outline of what is known and what has been tested.

Applications of Physics to Targetry

The basic science involved in cyclotron targetry is physics. The range of the charged particles in the target medium, the yields from the nuclear reaction and the density of the target medium all depend to a large extent on the physics of the situation.

The mathematical description of the ranges of charged particles in different media was first published by Bethe (1). The basic equation has not changed since then. There haven been a few terms added to the equations to account for small effects in the electronic and nuclear shell component to the stopping power. More recent work by Janni has resulted in an equation and computer program to calculate proton ranges in any element (2). The general equation is:

$$\frac{dE}{dx} = \frac{4\pi z^2 e^4}{m_0 V^2} \frac{Z}{A} \ln \frac{2m_0 V^2}{I}$$

Where I= ionization potential Z= atomic number

x= distance A= atomic mass

V= velocity $\quad$ z= particle charge

m_0= mass

This equation can also be used to give a good approximation to the range of a proton in any medium made up of more than one element by calculating an average Z and an average atomic weight for the medium and using these values in the equation. Ranges of particles other than protons can be determined by scaling the range for protons to that for other particles.

$$S_d(E)=S_p(E/2)$$
$$S_t(E)=S_p(E/3)$$
$$S_{He\text{-}3}(E)=4S_p(E/3)$$
$$S_{He\text{-}4}(E)=4S_p(E/4)$$

The result is that if the average density of the medium can be determined fairly accurately, then the range can also be calculated to the precision to which the density is known. The ranges of the particles can be increased by a factor of two by changes in fluid density encountered in the typical cyclotron target and beam currents. It very important that this density reduction be minimized in the design of a target.

The yield of a target at any point in time is given by the following equation.

$$\text{Yield} = SY(1-e^{-\tau t})$$

Where SY = Saturation Yield

τ = decay constant for the isotope

and t = time

The saturation yield is determined from the cross section measurement. This quantity is usually measured experimentally and reported in the literature. Sometimes it is necessary to do a numerical integration of these values to obtain the thick target yield since these values are not always experimentally determined. The saturation yield is the maximum attainable and is not usually obtained in routine cyclotron irradiations although under certain circumstances it is approached. Several numerical integration programs are available to get the saturation yield from the cross-section data with varying levels of sophistication. The data in most cases do not justify a very sophisticated integration routine and a simple one will suffice.

The phenomenon of density reduction is most obvious in gas targets. The density is very dependent on the temperature of the gas and the extent of ionization of the gas. The beam will of course travel farther in a less dense gas. This effect is not linear with beam current and can be a severe limitation to the use of high beam current targets.

Another major means of loss of isotope production yield is the phenomenon of multiple scattering. It is of major concern only in gas targets. As the beam passes through the target medium, it is scattered by the electron density of the atoms. This causes a small deflection in the beam direction and as a result the beam can be scattered into the target walls. A simple equation for the average angle of deflection is given below:

$$\langle \Phi^2 \rangle = (E_s/\beta pc)(x/X_0)$$

Where : $Es = (4\pi/\alpha)1/2mec2 = 21$ MeV

pc = momentum

β = velocity relative to light

x = thickness of matter (g/cm2)

X_0 = the radiation length

This equation can be rearranged to

$$\langle \Phi^2 \rangle = [21U/(U^2-(938.2)^2)]^2 x/X_0$$

Where : U = total energy (E+938.2 MeV)

x = range of the particle (g/cm^2)

X_0 = thickness of the material (g/cm^2)

In longer targets this scattering may result in the loss to the wall of half or more of the beam. It is usually necessary to minimize this loss by making the target as short as possible. This also means that a high pressure of gas must be used in gas targets in order to maximize the yields.

Application of Chemistry to Targetry

The are several situations where chemistry is involved in cyclotron targetry. The most obvious is the relationship to hot atom chemistry since this is the basis of the chemical reactions occurring in the target during irradiation. Data from hot atom chemistry gives us a place a to start but the conditions are quite different in the two cases. In the typical hot atom experiment, the beam current is 1 microampere or less while in the typical cyclotron target, the demand for high yields requires that beam currents of 15 to 30 microamperes be used. In lower energy cyclotrons, this current must be even higher to produce sufficient quantities of the radioisotope for the radiochemist to have a reasonable amount of the radiopharmaceutical for injection at the end of synthesis. Basically the same reactions are occurring in the two situations and the data from hot atom results has been used in the past to predict the chemical form of a radioisotope produced in a production target. The production of methane in the nitrogen-hydrogen target is based on the finding of the hot atom chemists who first studied the reactions of carbon atoms in the gas phase in the presence of hydrogen.

The other place where the chemistry is important is in predicting the relative yields of the radiochemical species as a function of the temperature of the surrounding gas. The temperatures in the beam strike area may reach 300°C and this can have an effect on the chemistry. The distribution of temperatures in an argon gas target is given in Figure 1.

Applications of Engineering to Targetry

The most serious problem encountered in cyclotron targetry is the removal of heat from the target during the irradiation. If the heat is not removed in an efficient manner, the temperature inside the target will rise and the yield from the target will fall. The choice of materials will depend not only on the chemical properties of the material, but also on the thermal properties. There is a constant flow inside both liquid and gas targets which help with the convective heat transfer and advantage should be taken of this flow in the design of the targets. The conditions under which this flow changes from laminar to turbulent is an area under study. This flow will have a profound effect on the heat transfer characteristics of very high beam current targets as are now being suggested for small, low energy accelerators.

Target foils are the most important aspect of target design and considerable attention must be paid to the thermal, chemical, and mechanical nature of the material used. There is not one perfect material and compromises must be made in choosing the best foil for a particular target. The stress placed on the foil is given by the equation:

$$\sigma = 0.396[P^2Ea^2/h^2]^{1/2}$$

Where : P= pressure (psi)

E= Young's Modulus

a= radius of foil (in.)

h= foil thickness (in.)

σ= stress on the foil (psi)

The yield strength of the foil as a function of temperature is sometimes a difficult property to find for a particular alloy such as HAVAR (Hamilton Metals, Lancaster PA.). The yield strengths of some metals commonly used for target foils versus temperature are given in Figure 2. This is an important factor in targets for low energy accelerators which require thin windows to maximize the energy but higher pressures in order to reduce the length of the target to minimize the loss of beam due to multiple scattering.

The exact design of the foil retaining ring on the target can have a profound effect on the ability of the foil to withstand stress. It has been shown that a slightly curved, bevelled retaining ring is very advantageous in minimizing the stress on the edges of the foil.

Design Considerations for Gas Targets

Targets utilizing a gas as the target material are perhaps the most sensitive to design criteria. They are also by far the easiest to use and are therefore the most popular. A flow chart for the design of a gas target is given in Figure 3. There are a number of factors which must be balanced against one another in order to decide on the best target design for a particular consideration. In the past, targets were designed without much in the way of calculating the optimum design. If they worked that was fine and if they didn't they were redesigned in order to make them work.

The primary consideration in the design of a target is the optimum beam entrance energy for a particular reaction. This is constrained by the cross-section of the reaction versus energy and the maximum practical energy of the cyclotron. If the reaction does not produce much activity at low energy, then the beam at an energy below this level should be dumped into the rear of the target in order to minimize the heating and density reduction in the target gas.

The next thing which must be determined is the foil thickness. This in turn will determine the energy incident on the gas. The foil should be thin enough so that there is not much energy loss and not much power deposited but thick enough to withstand the pressure of the target at elevated temperatures. The heat transfer characteristics of the foil must be taken into account so that the temperature of the foil is kept to a minimum.

The target must usually be water cooled. If the water cooling is inefficient, then the target will heat up and the gas will expand. When this happens either the pressure will be increased a great deal if there is not much dead volume in the target, or the density of the gas in the beam strike area will be reduced. There must also be helium cooling or air cooling on the foil of the target in order to maximize convective heat transfer from the foil. The target shape can also be modified to some extent. By far the easiest shape to machine is the cylinder. Some machinists will balk at the idea of producing a conical target. The conical shape is the best as far as minimizing the amount of target gas which must be used to produce a given amount of isotope. This is a result of the multiple scattering in the target. The beam density decreases as it passes through the gas and the angle of spread is determined by the thickness and density of the target foil as well as the gas. A radioisotope of higher specific activity will be produced in a conical target.

The topic of specific activity is of great concern to people who are trying to do receptor binding studies. The design of the target should try to minimize to amount of cold carrier which will be introduced by the target. This is especially important in the case of the carbon-11 target. The specific activity can be ruined by using a viton O-ring in an inappropriate place. The degassing of the viton can add a large amount of carrier carbon to the target gas. The purity of the target material is also of critical importance in this respect. The nitrogen gas available from various vendors can vary a great deal in purity and care should be taken to ensure that the target gas is as free from carrier as is possible. Analyzed gas will often be of higher purity than unanalyzed gas of similar stated purity.

The last consideration is whether the target should be run in batch mode or in a continuous mode. The batch mode has the advantage that the amount of target material used is kept to a minimum while the continuous mode has the advantage of letting one know exactly where one stands with respect to the production of the radioisotope. If the target is run in the continuous mode, it is possible to continue the beam a little longer if the production seems low where this is not possible in the case of the batch target.

Design Considerations for Liquid Targets

The considerations for liquid targets are somewhat different than for gas targets. The problem of multiple scattering is much less severe since the density of the medium is so much higher and therefore the path length of the beam is so much shorter. The main problem associated with the liquid target is to keep the liquid from boiling and thereby reducing the average density. One of the easier methods to increase the boiling point of the liquid is to increase the pressure over the liquid (ref Heselius et al.). The heat transfer out of the liquid can also be increased by recirculating the target liquid. This can be done by pumping the liquid through the target.

Since pressures in liquid targets are not usually as high as those in gas targets, the foils in general can be thinner and thus the energy higher. The range of the particles can be predicted more accurately in liquid targets since the problem of density reduction in the target is less severe if the liquid is not boiling. This allows for an accurate prediction of the thickness the target needs to be in order to gain the maximum from the cross-section and dump the less useful lower energy part of the beam into the rear wall.

Design Considerations in Solid Targets

Solid targets have there own problems in that some materials of use in producing short-lived positron emitting isotopes are nor efficient conductors of heat. This can cause problems in the vaporization of the target material. The deposition of heat is confined to a much smaller area and this can cause temperatures to increase to a point where the material is vaporized. The other major inconvenience with solid targets is that the material is sometimes difficult to recover from the target. In many cases the entire solid must be processed and the radioisotope of interest extracted. There is also a much greater chance for contamination since solids often adsorb gases from the surrounding atmosphere at low temperatures. These gases may then be released when the beam is applied or may combine with the desired radioisotope to produce undesirable chemical species.

The removal of heat from the solid target is in general easier since the cooling medium can be in close proximity to the target material. It is also usually possible to use convective heat transfer by flowing a stream of inert gas either over the target or through it. In fact this gas stream has been utilized to remove the radioisotopes from the target material in the form of gaseous molecules.

Target Modeling

During the past few years computer programs have become available to model some of the parameters in target design. It is now possible to calculate foil temperatures given the some parameters relating to the three modes of heat transfer, the beam current and assuming a uniform beam distribution. The more accurate model using the actual beam distribution has not been implemented but is certainly feasible. It is possible to know fairly accurately what the equilibrium temperature of the foil will be under a given set of circumstances. This in turn allows predictions to be made regarding the maximum stress the foil can withstand.

The cross-section of a nuclear reaction can be calculated with fair accuracy but for low Z materials the experimental measurements are by far the more accurate. There is good data on all the usual nuclear reactions used to produce the short-lived positron emitting isotopes. Almost any numerical integration routine may be used to determine the thick target yields from the cross-section data.

Heat transfer within the target is a much more difficult problem to calculate sine often the exact distribution of the beam inside the target is not known due to the effects of density reduction and multiple scattering. It is possible to make some estimates using very simple mathematical approximations.

Yield strengths of materials must be determined from experimentally determined values. The mathematical models are simply too complex for use on a small computer and result in answers which are not accurate enough. Data are available for most common materials.

The small angle multiple scattering in the target can be approximated relatively easily from the formulae given above. The answers obtained from simple models are in general sufficient for target design since the exact distribution of the beam inside the target is not usually known.

Conclusion

It is possible to design a target which will produce the desired results on the first attempt if all the available information is utilized. Unfortunately this is not usually the way things are done since people must deal with such mundane matters as time constraints, available materials and cranky machinists. In the future it should be possible to combine the collective knowledge gained by people over the years to produce efficient and reliable targets.

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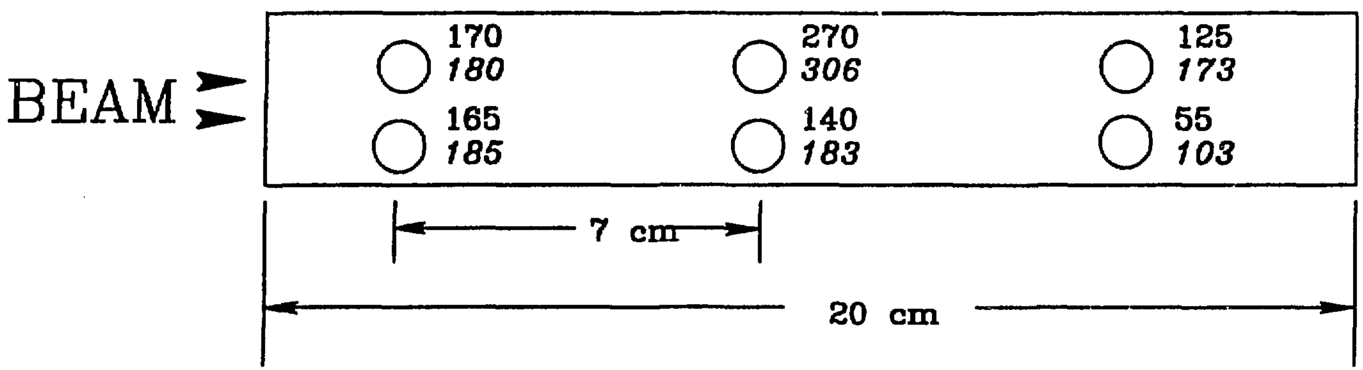
Figure Legends

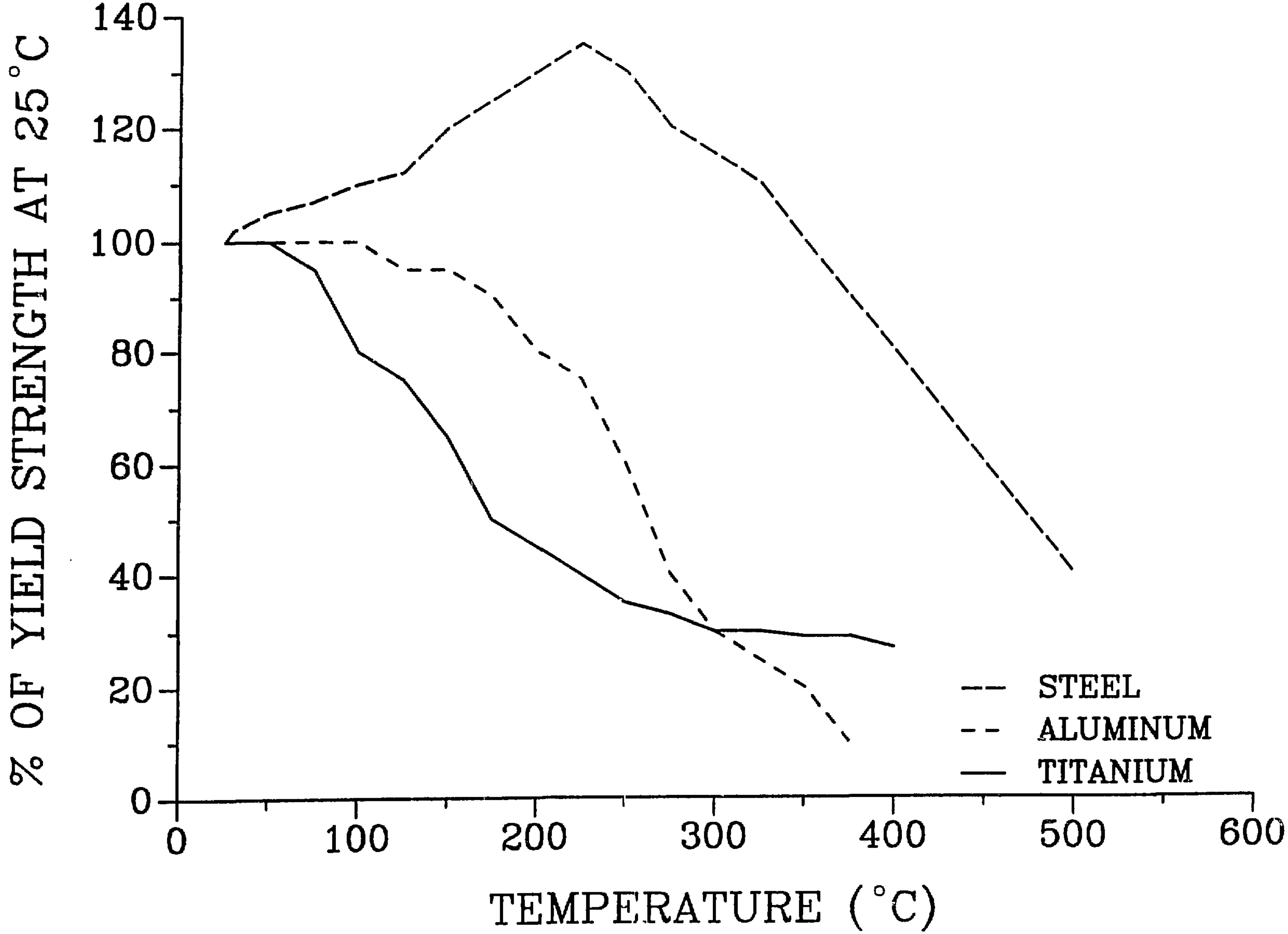
Figure 1. The temperature distribution inside an argon gas target with a 14.7 MeV beam of protons at a current of 15 μA incident on the gas. The upper numbers are at a pressure of 675 kPa and the lower numbers at a pressure of 550 kPa.

Figure 2. The yield strength of foil materials with a change in temperature. The yield strength is given as the percentage of the yield strength at 25°C. The temperature is given in degrees centigrade.

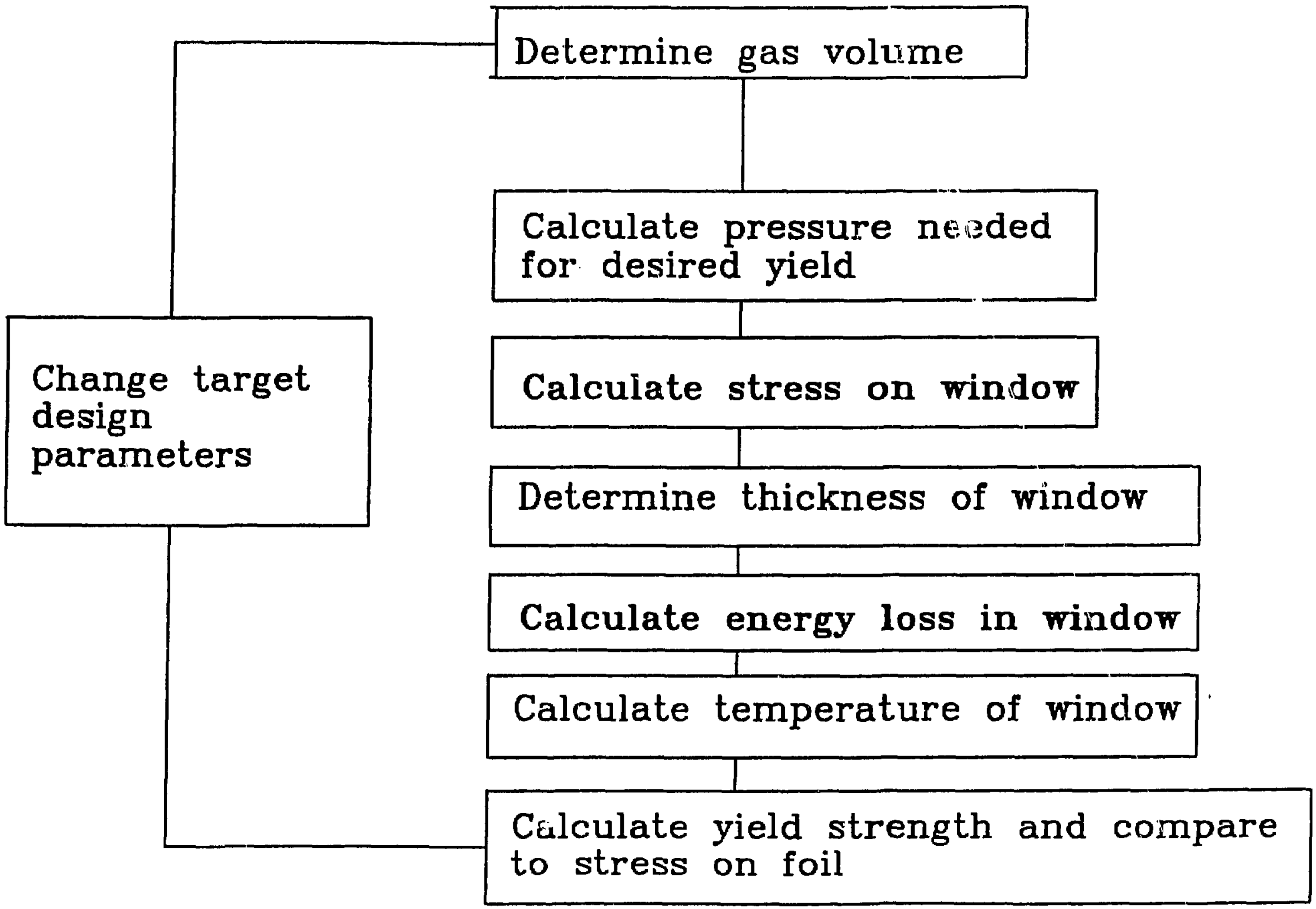
Figure 3. Flow chart of target design. The is no final solution, only a series of compromises to meet the specific application of the target.

Target Temperatures





% OF YIELD STRENGTH AT 25°C
140
120
100
80
60
40
20
0
0
100
200
300
400
500
600
TEMPERATURE (°C)
STEEL
ALUMINUM
TITANIUM


Determine gas volume
Calculate pressure needed for desired yield
Calculate stress on window
Determine thickness of window
Calculate energy loss in window
Calculate temperature of window
Calculate yield strength and compare to stress on foil
Change target design parameters