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## MARANGONI EFFECTS IN THE BOILING OF BINARY FLUID MIXTURES

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### ABSTRACT

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Results of very recent experimental studies indicate that during nucleate boiling in some binary mixtures, Marangoni effects augment the gravity driven flow of liquid towards the heated surface. With gravity present, it is impossible to separate the two effects. The reduced gravity environment gives an unique opportunity to explore the role of Marangoni effects on the boiling mechanisms free of gravitational body forces that obscure the role of such effects. However, recent experimental results suggest that under reduced gravity conditions, Marangoni effects is the dominant mechanism of vapor-liquid exchange at the surface for some binary mixture.

To further explore such effects, experiments have been conducted with water/2-propanol mixtures at three different concentrations under normal gravity with different orientations of the heater surface and under reduced gravity aboard the DC-9 aircraft at NASA Lewis Research Center. The system pressure was sub atmospheric ( $\sim 8$  kPa at  $1g_n$ ) and the bulk liquid temperature varied from low subcooling to near saturation. The molar concentrations of 2-propanol tested were 0.015, 0.025 and 0.1. Boiling curves were obtained both for high gravity ( $\sim 2g_n$ ) and reduced gravity ( $\sim 0.1g_n$ ). For each concentration of 2-propanol, the critical heat flux has been determined in the flight experiments only for reduced gravity conditions. Comparison of boiling curves and CHF obtained under  $1-g_n$  and reduced gravity indicates that boiling mechanism in this mixtures is nearly independent of gravity. The results also indicate that the Marangoni mechanism is strong enough in these mixtures to sustain the boiling under reduced gravity conditions.

### INTRODUCTION

In many technological applications, vaporization of a working fluid is critically important. It facilitates the heat input in Rankine power systems, the cooling effect in vapor compression heat pumps and high heat flux removal in thermal control applications. When the working fluid is a pure substance, as is often the case, the characteristics of the vaporization process can be predicted with reasonable accuracy using the results of extensive research on boiling of pure liquids over the past 60 years. Recent efforts to improve component or system performance have lead some developers to consider the use of binary working fluids. In general, researchers have found that some binary mixture working fluids offer the potential for improved thermodynamic efficiency, or superior heat transfer performance. The advantages of using binary mixture working fluids in heat pump systems have been explored by several investigators (see, for instance, Domanski (ref. 1)). This type of investigation invariably points out the fact that the current limited capability to accurately predict the transport processes for boiling of binary mixtures is a major obstacle to the development of heat pump systems using binary working fluids. Another perspective on the significance of Marangoni effects during binary mixture boiling can be obtained by considering two-phase thermal management systems for spacecraft. Systems of this type using pure working fluids have been extensively studied by Degroff et al.(ref. 2) and others.

Results of some very recent studies suggest that it may be possible to enhance the ability of the system to resist dryout and/or the onset of film boiling by using a binary coolant than a pure working fluid. McGillis and Carey (ref. 3) experimentally examined the pool boiling of mixtures of water and alcohol at low pressure on a small, 1.3 cm square heated surface. Boiling curves and the critical heat flux were determined for several different alcohol-water mixtures at a number of pressure and concentration combinations. Methanol/water, 2-propanol/water and ethylene glycol/water mixtures were tested. These experiments were done in a closed thermosiphon system so that concentrations in the liquid pool can be accurately maintained at a predetermined level throughout the test. At low concentrations of 2-propanol in water, these investigators have found that the critical heat flux (CHF) may be enhanced above that for either of the pure fluid components under comparable conditions. Interestingly, at low concentrations of ethylene glycol in water, the critical heat flux is observed to be lower than that of water. McGillis and Carey (ref. 3) found that the variation of the critical heat flux with concentration correlates strongly with the surface tension gradient or the Marangoni effect. 2-propanol/water is a positive mixture where the more volatile component has a lower surface tension than the surface tension of the less volatile component and the surface tension

gradients arising from the preferential evaporation of the more volatile component at the heated surface act to enhance the liquid motion towards the surface. Conversely, ethylene glycol/water is a negative mixture where the surface tension gradients act to decrease the liquid motion towards the heated surface. Beginning with the well-known Zuber correlation for the critical heat flux for an upward-facing flat heated surface, these investigators replaced the restoring effect of buoyancy in the Zuber correlation with the combined effect of buoyancy and surface tension gradients. McGillis and Carey (ref. 3) found that by adjusting a constant in the correlation, they could match critical heat flux data for both their water alcohol mixtures as well as the ethanol and water data of Reddy and Lienhard (ref. 4). Agreement is quite good over the entire range of concentrations tested, and this model correlates these data better than any other currently available schemes.

The important point here is that for certain binary fluid mixtures, there is abundant evidence that surface tension gradients resulting from concentration differences act to enhance the fluid motion towards the heated surface. For some mixtures the effect on the critical heat flux is so great that this effect appears to be substantially stronger than the normal buoyancy effect at  $1-g_n$ . It is difficult, however, to fully gauge the strength of the Marangoni effect because it is always combined with buoyancy. By conducting binary mixture nucleate boiling studies under reduced gravity conditions, the buoyancy effect would be removed, and the ability of Marangoni forces to induce liquid motion towards the surface would be directly observable. In addition, reduced gravity binary boiling experiments may also pave the way for the use of binary coolants in spacecraft thermal control applications. The present study is intended to investigate the Marangoni mechanism in the boiling of binary mixtures in a reduced gravity environment. Boiling of 2-propanol and water mixtures at three different concentrations have been investigated in a DC-9 reduced gravity aircraft which provides 20-25 seconds of reduced gravity. The gravity level attained by the aircraft is about  $0.01g_n$ .

Siegel and Usiskin (ref. 5) conducted the first reduced gravity boiling experiment in a 0.7-second drop tower. Siegel (ref. 6) published a comprehensive summary of the early studies up to the mid-1960s. Recently, the effects of variable gravity on boiling heat transfer were summarized by Merte (ref. 7). Straub et al. (ref. 8) conducted microgravity boiling experiments for more than 15 years. Most of the studies conducted under reduced gravity or variable gravity were for pure liquids. However, Abe et al. (ref. 9) recently conducted pool boiling experiments with water-ethanol mixtures under microgravity conditions. These investigators found that the heat transfer was enhanced under microgravity and they ascribed this enhancement to the Marangoni flow due to surface tension gradient.

## SYMBOLS

°C	Degree centigrade	CHF	Critical heat flux	g	Instantaneous Acceleration
$g_n$	Normal gravitational acceleration	P	Pressure	$q''$	Heat Flux
t	Time	$X_p$	Molar concentration of 2-propanol		

## EXPERIMENTAL SETUP AND PROCEDURE

The test section used in our experiments is a 12" long square (3") channel made of stainless steel. The top plate and the two side plates are 1/4" thick and the bottom plate is 1/2" thick. The bottom plate has a rectangular cut-out to accommodate replaceable heated surface element. The heater element is made of oxygen-free, high purity copper to ensure that the thermophysical properties of the element are defined to high accuracy. The copper heating element is silver soldered to the stainless steel holder. The contact area between the stainless steel and the copper is minimized to avoid heat loss. The heat loss is computed from a simple model and it is less than 5% of the heat input. Electric cartridge heaters fitted into the bottom of the copper element provide the heat input which flows along the bar of circular cross section to the flush end exposed to the flow in the test section. The flush end of the copper heater element is a 1.2 cm diameter circular finger. Miniature thermocouples installed along the copper bar allow measurement of the temperature gradient in the bar, and hence the heat flux to the surface exposed to the flow. Lateral side walls of the test section have rectangular windows made of transparent polycarbonate for flow visualization. The system pump delivers fluid to the inlet header of the test section. A porous plate in the inlet header helps provide even flow distribution in the test section. Flow exiting the test section is piped to the system condenser. A Validyne pressure transducer installed in the test section is used to monitor the pressure in the system. During reduced gravity flight test, a high speed video camera was mounted in front of the window of the test section for visual recording of the boiling process. Figure 1 shows the layout of the experimental hardware.

The experimental setup consists of two flow circuits; one for the binary mixture and the other for the coolant flow in the condenser. The components of the binary mixture circuit are the test section, a pump and the tubes in the condenser. The heater element at the bottom of the test section heats up the binary mixture. The purpose of the pump is to maintain a constant flow in the test section and the condenser. The intention of this design is to provide a weak bulk convective motion that will not affect the nucleate boiling process on the heated surface, but will carry vapor bubbles that leave the surface during the boiling process to the condenser. The objective is to sustain a steady nucleate boiling process while maintaining constant pressure and bulk concentration in the test section. The coolant circuit consists of coolant pump, a coolant tank and the shell of the condenser. A support structure for the test system is built by using 13/16" aluminum Unistrut bar. The test system along with its support structure is mounted in one of two Learjet racks provided by NASA. The computer, data acquisition system and all the electrical components are mounted on another Learjet rack. The thermocouple and pressure data are monitored using a PC-based data acquisition system.

The binary mixture circuit is evacuated by a vacuum pump. The fill port is opened and the binary mixture flows from the charging tank to fill the system. The pressure of the system can be lowered by bleeding vapor from the evacuation tank or can be increased by filling the system with more liquid mixture. When pressure becomes steady, the pumps are turned on. Electrical power is supplied to the cartridge heater and the heat flux to the binary mixture is controlled by a variac. The heat flux is obtained from a least-square fit of the five thermocouples embedded along the heater element and the surface temperature is computed by extrapolation. The experiment is continued until the system reaches the critical heat flux.  $1-g_n$  experiments were conducted at two different heater surface orientations, upward facing and downward facing. At first, the system pressure and temperature are stabilized for the upward facing heater surface and the experiment is conducted for this orientation. At the end of the experiment, there is a cool down period when the system pressure and temperature are re-stabilized. At this point, the rack is oriented to conduct experiments for the downward facing configuration. During the flight experiment, pressure was maintained constant in each flight experiment. At the beginning of every other parabola, the heat flux was set and the temperature profile in the heater element was monitored and recorded. When the flight experiences a transition from  $2g_n$  to reduced gravity, the gravitational part of the heat flux is expected to approach zero while the heat flux due to Marangoni effect becomes the dominant mechanism to sustain nucleate boiling. Therefore the system experienced a transient heat transfer mode and restabilized at a new heat flux value which can be monitored from the thermocouple readings. The normal gravity experiments for the boiling of binary mixture were performed in the Multiphase Transport Laboratory at the University of California at Berkeley and the reduced and elevated gravity experiments were performed in the DC-9 reduced gravity aircraft of NASA Lewis Research Center.

## RESULTS AND DISCUSSIONS

Figure 2 shows the boiling curve for 2-propanol/water binary mixture at two different orientations of the heater surface; upward facing and downward facing. The molar concentration of 2-propanol in the mixture is 0.015. Also shown in the plot are the critical heat fluxes for water at same orientation and same system pressure. It is evident from the plot that the critical heat flux of 2-propanol/water ( $X_p=0.015$ ) is greater than that of pure water under the same condition by a factor of three at same system pressure and same orientation. Another interesting point to note is that the critical heat flux of 2-propanol/water at downward facing heater configuration is substantially more than that of pure water at upward facing configuration. The trends of this normal gravity curve clearly suggests that there is additional mechanism in the boiling of 2-propanol/water mixture which is responsible for this higher critical heat flux.

The transient wall superheat and the  $g$ -level during an experimental cycle in DC-9 reduced gravity flight are shown in figure 3. The molar concentration of 2-propanol in water was 0.015 and the heat flux increased from  $89 \text{ W/cm}^2$  to  $98 \text{ W/cm}^2$  during this experimental cycle. Although the DC-9 aircraft is used for reduced gravity environment for 20-25 second, there is an elevated gravity ( $1.8g_n \sim 2.0g_n$ ) part during each parabolic maneuver which also lasts 20-25 seconds. During this change in gravity level, the hydrostatic pressure of the system changes by 2 kPa/ $1g_n$  and causes a shift in the saturation temperature of the binary mixture by  $4 - 5^\circ\text{C}/1g_n$ . The wall superheat changes during this change in gravity level but reaches a steady state value within  $1^\circ\text{C}$  during both elevated gravity period and reduced gravity period. Therefore, the data represented in the boiling curves are essentially steady state value for the system.

The temperature profiles along the heater were studied in detail to ensure the steady state condition of the system. Figures 4 and 5 show the transient temperature profile along the copper heater element obtained from five thermocouples embedded in the element for the same condition as in figure 3. The transient temperature profile during

low g to high g transition is shown in figure 4 where the system reaches steady state at a higher gravity level (i.e. higher saturation temperature) by establishing a higher wall temperature. Interestingly, the heat flux stays almost constant during this transition. The system moves towards a lower wall temperature during high g to low g transition as is evident in figure 5. However, the heat flux changes slightly from  $94.8\text{W/cm}^2$  to  $98\text{W/cm}^2$ .

The boiling curves at reduced gravity for three different concentrations are plotted in figure 6. The molar concentrations of the 2-propanol/water binary mixtures are 0.015, 0.025, 0.1 and the surface tension gradients of these mixtures decrease with concentration. The log-log plots show that the boiling curves roughly follow the general  $1/3$  power law. The boiling curves also show that the higher the surface tension gradient the higher the critical heat flux. It is evident here that the critical heat flux correlates strongly with the surface tension gradient and hence the Marangoni effect. At the heater surface, alcohol evaporates preferentially which has a lower surface tension. Therefore, the surface tension of the liquid close to the solid-liquid-vapor interface is higher than the bulk surface tension around liquid-vapor interface. This surface tension gradient apparently causes more liquid to flow towards the heater surface and delays the onset of dry-out. Figure 7 shows that the boiling heat transfer was virtually the same under reduced gravity and terrestrial conditions. However, the critical heat flux of the mixture under reduced gravity decreased by about 10% for 0.015 mole fraction mixture compared to the terrestrial level. Similar reduced gravity boiling characteristics of binary mixtures were reported by Abe et al. (ref. 9)

## CONCLUDING REMARKS

The data obtained in this investigation imply that the Marangoni effect arising from the surface tension gradients due to concentration gradients is an active mechanism in the boiling of binary mixtures such as 2-propanol/water. At a molar concentration of 0.015 of 2-propanol in water, where the surface tension gradient is highest among the concentration tested, the critical heat flux is a factor of three greater than that of pure water for similar conditions under normal gravity. Comparing the results of normal gravity and reduced gravity of the order of  $0.01g_n$ , it is found that the boiling curves and the corresponding critical heat fluxes are almost independent of the gravity level. The effect of gravity on the boiling curves of binary mixtures found in this investigation are consistent with that reported by Abe et al. (ref. 9)

## ACKNOWLEDGMENT

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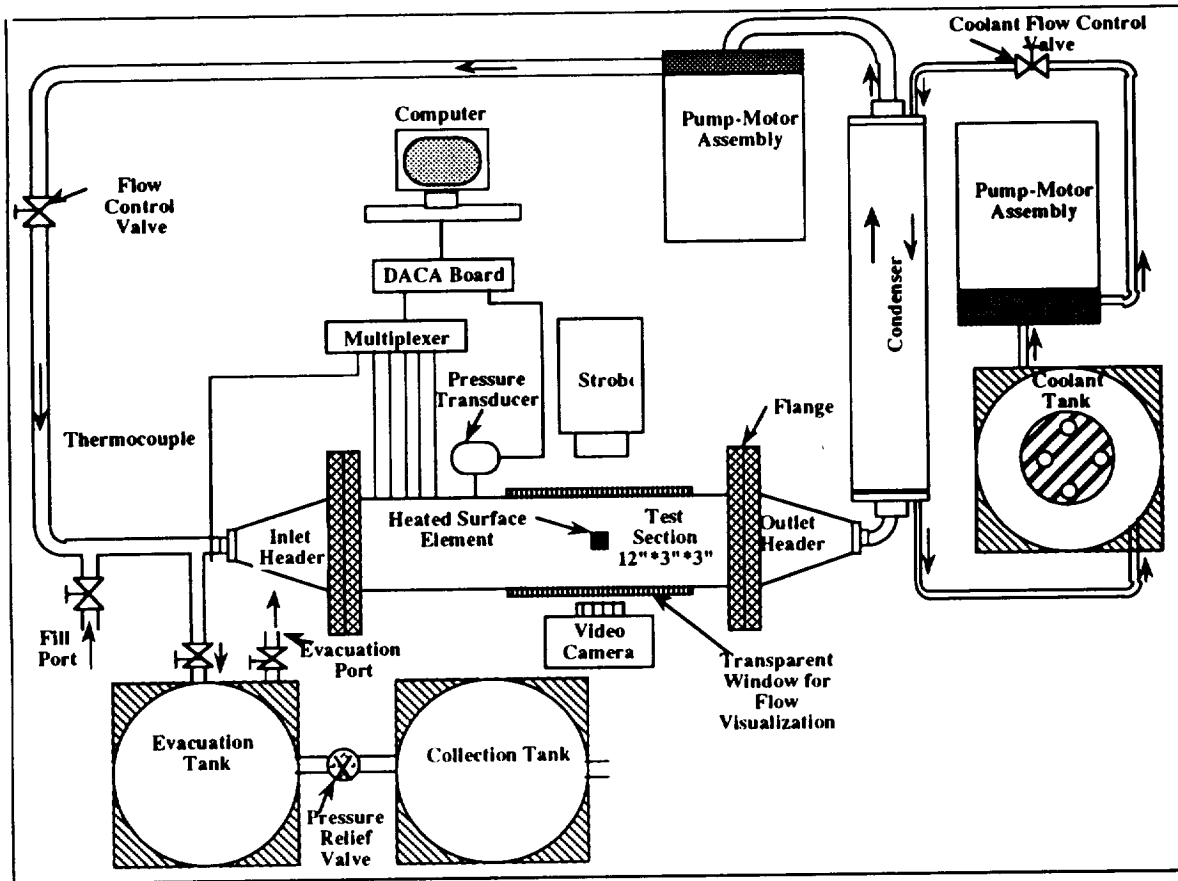


Figure 1.- Layout of the experimental system.

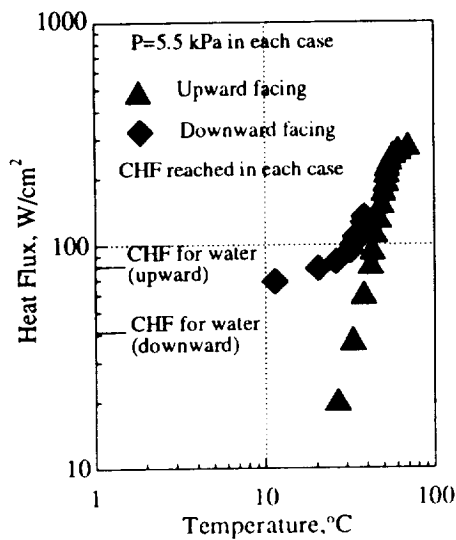


Figure 2.- Boiling curves for 2-propanol/water mixture ( $X_p=0.015$ ) at different orientations.

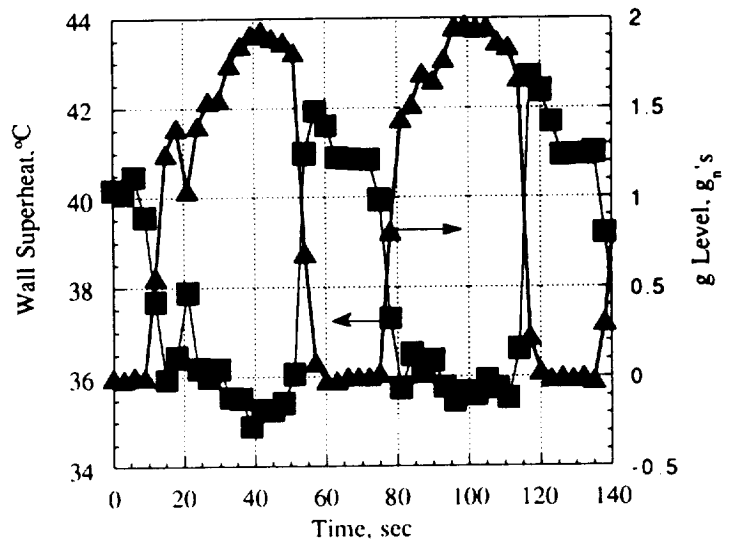


Figure 3.- Transient wall superheat and g-level during an experimental cycle for  $X_p=0.015$ .

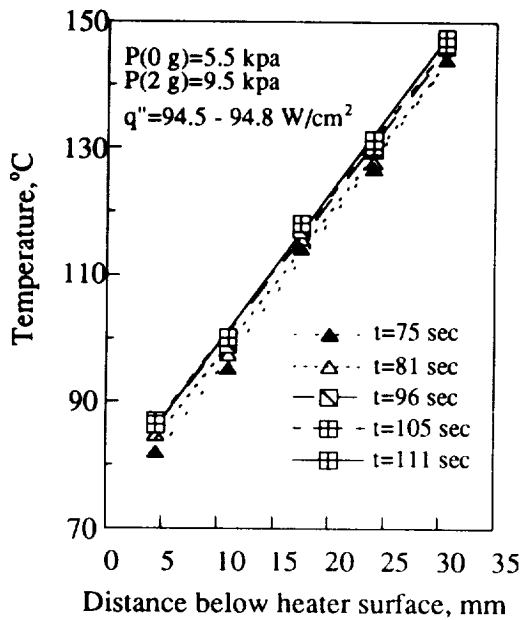


Figure 4.-Transient temperature profile along the heater during low g-high g transition for  $X_p=0.015$ .

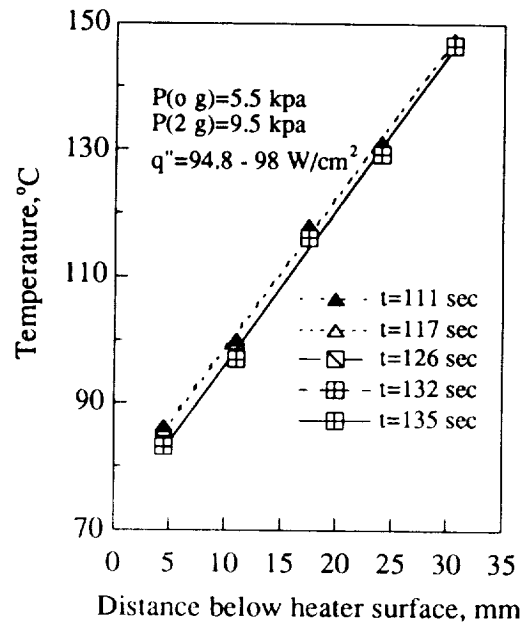


Figure 5.- Transient temperature profile along the heater element during high g-low g transition

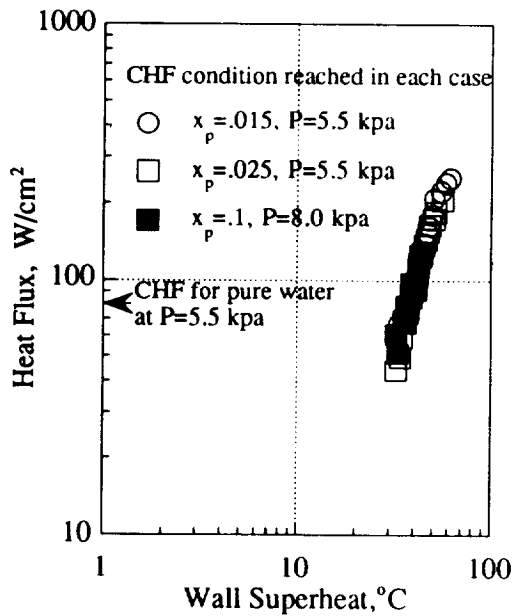


Figure 6.- Reduced gravity boiling curves for various concentrations of 2-propanol

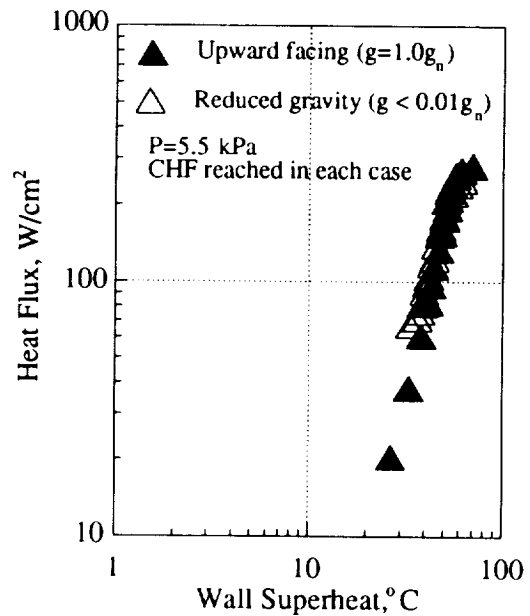


Figure 7.- Comparison of boiling curves between reduced gravity and terrestrial gravity