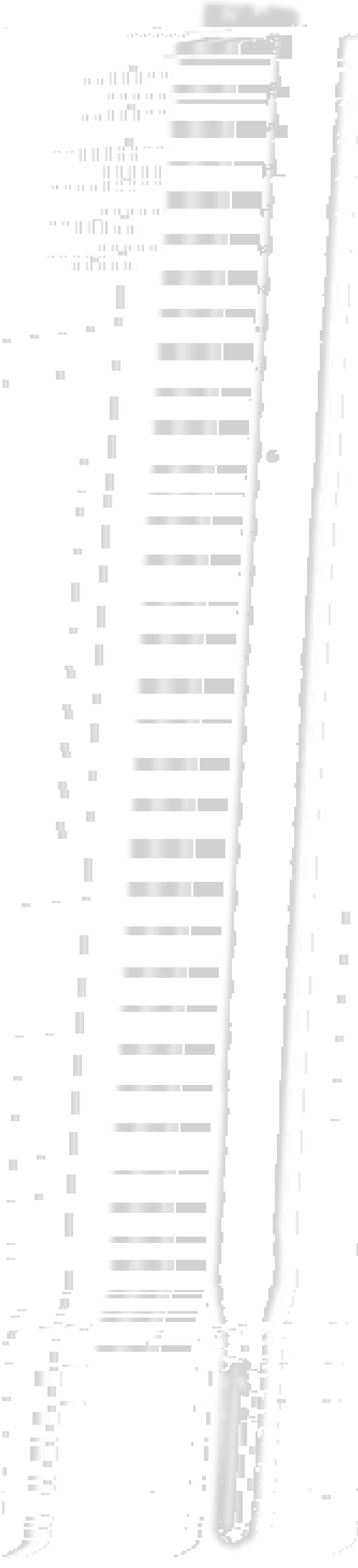
harmaceutical, biotechnology and specialty chemical companies are challenging the heat transfer community to provide solutions that enable critical processes to operate at extremely cold temperatures. In the past, it was adequate to operate at temperatures as low as -80˚F (-62.2˚C). Now industry continues to push for colder temperatures. Low-temperature heat transfer fluid manufacturers and heat transfer companies are being asked to provide systems that can run reliably at -148˚F to -184˚F (-100˚C to -120˚C).

Why such low temperatures? For certain chemical reactions the rule of thumb is that the reaction time is increased by a factor of two for each 18˚F (10˚C) reduction in operating temperature. If the temperature is too high, the reaction time is very quick, adversely impacting quality and repeatability of results.

A number of design considerations must be taken into account when operating at these extreme conditions. This article reviews the outcome of recent research of heat exchanger design and heat transfer fluid performance for low-temperature operation. It defines practical low-temperature operation of the various heat transfer fluids for a given type of heat exchanger. The performance characteristics of the different fluids are discussed, as is the performance of heat exchangers as heat transfer fluids begin to freeze within them.

Common low-temperature applications in a pharmaceutical plant are reac-



transfer fluid flow rates and inlet temperatures, as well as liquid nitrogen flow rates and operating pressures.

Heat transfer fluid properties

A good heat transfer fluid for low-temperature service must have a low freeze-point temperature, low viscosity and low thermal diffusivity. Depending on the operating range of the temperature control system, it might need to be capable of operating safely at hot temperatures. Table 1 compares the fluid properties of the four heat transfer fluids tested at -130˚F (-90˚C).

A generalized heat transfer correlation for the heat transfer fluid that defines how fluid properties impact heat transfer is expressed by:

Where:
Re = Reynolds number
Pr = Prandtl number
D_h = hydraulic diameter
C = constant
a = positive exponent that is less than 1.0
b = positive exponent that is less than 1.0

Key observations from the generalized correlation include:

- A high density, specific heat and thermal conductivity are good for heat transfer.
- A low viscosity is good for heat transfer.

It is important to keep the boundary layer thin, with efficient heat transfer through the boundary layer. The first bracketed expression is the Reynolds number, which is an indication of the thickness of the fluid boundary layer near the heat transfer surface. A high Reynolds number is important.

The second bracketed expression is the Prandtl number, which affects the temperature gradient through the boundary layer. It is an indication of the rate by which heat is given up by the heat transfer fluid to the coolant.

Table 2 shows just how different the Reynolds and Prandtl numbers can be in the case of 15,000 pounds per hour (lb/hr.) (6,800 kilograms per hour [kg/hr.]) of heat transfer fluid at -130˚F (-90˚C) in a heliflow heat exchanger. The exchanger has 12.3 square feet (sq. ft.) (1.14 square meters [sq. m.]) of heat transfer surface.

Testing showed that an evaluation of the freeze point of a heat transfer fluid is insufficient to determine the suitability of a fluid for a given application. The freeze point, along with the fluid properties, the heat exchanger design and fluid velocity, plays a part in inhibiting the onset of freezing. Just because the temperature of the fluid leaving a heat exchanger is well above the freeze point of that fluid does not necessarily mean the fluid will not freeze inside the heat exchanger. This consideration is key.

How each fluid performed

Freeze-point temperature alone is not an indicator of whether or not a fluid will freeze in a heat exchanger. Fluid properties, velocity and heat exchanger design play important roles as well. Fig. 1 and Fig. 2 compare the four test fluids under identical operating conditions.

Thermocouples attached to the heat transfer surface indicated whether or not the fluid was freezing onto the heat transfer surface. Note how dramatically different the thermocouple-measured temperature was when freezing occured. Once solid deposits are present, they act as an insulator and drive the surface temperature to much colder levels. The difference is more than 100˚F (56˚C) between unfrozen and frozen heat transfer surfaces for the conditions tested.

Even if the outlet temperature from the heat exchanger is well above freezing, deposit buildup can occur inside a heat exchanger. This effect can be insidious, as runaway freeze up can sneak up on the control system if it is not properly configured.

The performance graph for Syltherm XLT shows that one of the thermocouples indicates the presence of frozen deposits even when the outlet fluid temperature is -90˚F (-68˚C). Syltherm XLT freezes well below -90˚F, at -168˚F.

As the Syltherm XLT is progressively cooled to approximately -100˚F inlet and -110˚F outlet, another region in the heat exchanger experiences a condition of freezing and defrosting. When the fluid is cooled further to -110˚F inlet, that region freezes entirely.

The freezing and defrosting condition

is imperceptible by a control system monitoring heat transfer fluid outlet temperature. The freezing/defrosting condition occurs because as the ice begins to form, local velocity increases, increasing the local heat transfer coefficient and changing the temperature distribution near the ice surface. A warmer condition is created at the deposit surface, melting the deposit. Not until the fluid temperature is lowered is the temperature differential sufficient to overcome the defrosting condition.

Take note of how different the thermocouple-measured temperature is between an unfrozen and frozen condition. The thermocouple measures a temperature at the heat transfer surface of approximately -150˚F when deposits are not present.

When a deposit is formed, it represents a step change in temperature. The temperature drops very quickly to approximately -275˚F. The solid is an insulator. Warm heat transfer fluid cannot readily conduct heat through the deposit. Therefore, the surface temperature under the deposit approaches the

exchanger performance is via thermocouples attached to the heat transfer surface; however, not all heat exchangers lend themselves to such a setup. When a thermocouple senses a dramatic drop in temperature, say 100˚F or more, freezing already is occurring at that location. Liquid nitrogen flow rate can be lowered momentarily until the deposit is driven off by the change in temperature gradient that will result.

Lower liquid nitrogen flow rate reduces the heat transfer coefficient on the coolant side, warming the heat transfer surface. This type of proactive control is excellent and far better than other reactive methods. It does not impact heat rejection by the exchanger. Normally, one or two minutes of reduced