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24 July 2004

**Abstract:**

**BETAÏNE BASED HEAT TRANSFER FLUIDS AS A NATURAL SOLUTION FOR ENVIRONMENTAL, TOXICITY AND CORROSION PROBLEMS IN HEATING AND COOLING SYSTEMS**

**The problem and importance**
The reason for this R&D effort was the environmental risk and toxicity of many conventional heat transfer fluids like glycols. Fluid toxicity and environmental risk in building systems and industrial use is becoming increasingly less acceptable. Alternative fluids have been sought from among salt solutions and vegetable oils. Technical difficulties have made broad scale use of these alternatives not very attractive. Main technical problems have been high viscosity and corrosion.

**The work**
A new possible alternative, a betaïne-water solution was tested and developed for heat transfer use. Betaïne (trimethyl glycine) is a by-product of the sugar industry and is acquired from sugar beet. Therefore betaïne is completely non-toxic and also non-harmful to the environment. Betaïne is a renowned substance in the pharmaceutical-, cosmetic-, food- and medical industry. Adequate freezing protection is achieved with betaine concentration of 20-55 % by weight. This solution was found to have other good qualities also; it has good heat transfer abilities, is not harmful to materials used in systems and has natural corrosion protection properties.

The development work consisted of carefully measuring freeze protection and pipe burst protection with different betaïne concentrations, measuring physical properties in same circumstances and developing the additive package for enhanced corrosion protection. Field-testing was done during four years.

**The conclusions**
Testing showed that a betaïne-water solution has good thermodynamic properties to be used as a heat transfer fluid in the HVAC industry, with additional environmental-, non-toxicity- and corrosion-properties. The concrete result was a trademark and application patent protected new product for the industry.

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**Paper:**

**BETAÏNE BASED HEAT TRANSFER FLUIDS AS A NATURAL SOLUTION FOR ENVIRONMENTAL, TOXICITY AND CORROSION PROBLEMS IN HEATING AND COOLING SYSTEMS**

## INTRODUCTION

The transfer of heat is commonly required in industry, buildings and municipal engineering systems. While the advances made in heat transfer technology and equipment have been great, the potential and benefits provided by a careful choice of the heat transfer medium are often overlooked. The selection of the right fluid offers savings in materials and running costs and can therefore significantly affect the life cycle cost.

The most important technical characteristics of a heat-transfer fluid are freezing protection, thermal and microbiological stability, physical properties which ensure good heat transfer properties, and low induced corrosion.

The increasingly stringent environmental legislation and public attitude call for improved environmental performance and non-toxicity. Consequently, development efforts focus on new types of heat transfer fluids that are environmentally friendly and less toxic than current solutions but possess excellent cold flow characteristics and heat transfer properties.

The main environmental characteristics of a heat transfer fluid are that it has a low environmental impact, that it is non-toxic, safe to use, and easy to dispose of or recycle, and that the total costs over the product's life cycle are competitive.

The starting point for this R&D effort was the toxicity problem of many conventional heat transfer fluids. Fluid toxicity in building systems and industrial use is becoming increasingly less acceptable. Alternative fluids have been sought from among salt solutions and vegetable oils. Technical difficulties have not made extensive use of these alternatives very attractive. Main technical problems have been high viscosity and corrosion.
The research work focused on Betaine (Trimethyl glycine or Glycinebetaine or TMG, see Figure 1) and its possibilities for avoiding existing problems in conventional glycol and salt solution use. Chemically betaine is a quaternary ammonium compound with three methyl groups attached to the nitrogen atom of a glycine molecule.

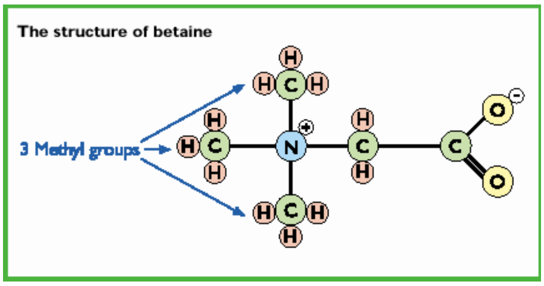


Figure 1: The structure of Betaine (Trimethyl glycine)

The Betaine used in this research and development work is fully natural and is acquired from sugar beet, the most common source for betaine (1). Conventional uses of Betaine include animal feeds, human food supplements, pharmaceuticals (2) and cosmetic products.

## 1 BETAINE TECHNICAL PERFORMANCE RESEARCH

The quality of a heat transfer fluid is defined by its technical performance, environmental aspects and economical impact. Technical performance was evaluated by measuring several different physical properties of betaine-water solutions and by conducting field tests in actual HVAC systems (heating, ventilation, air conditioning and refrigeration). Most measurements were done in or for the Fortum Technology Center, Porvoo, Finland.

### 1.1 Freeze Protection and Burst Protection

The most basic function of a heat transfer fluid is freeze protection. Due to low ambient temperatures or low operational temperatures of systems it is quite common for HVAC systems and industrial processes to require heat transfer in sub-zero conditions. In the operational temperature range fluids must remain in liquid form and not damage equipment by freeze-expansion (burst protection). The phase change chemistry of heat transfer fluids can be very problematic. With water it is easy to determine the freezing point. Phase change is immediate and self-evident.
When antifreeze agents are used the phase change becomes less evident, a non-stable area can be observed. For technical reasons two interesting temperature points are measured: the freezing point and the congealing point.

The freezing point is the temperature below which solid crystals may build up in a solution. The method used for freeze point determination in this study is the ASTM D 2386-97. To determine the freezing point, a solution is cooled down while stirring, until crystals appear. After the first crystals have built up, the solution is left to warm up slowly while stirring. The temperature in which all the crystals have disappeared is the freezing point of the solution.
At the freezing point, and at least 4 ºC below this point, the fluid is still functional, but the viscosity starts to climb rapidly. This is the non-stable area, where crystallization can appear suddenly. It is most common that the freezing point value is given for heat transfer fluids according to this method.

The congealing point is the other interesting point. The method used for congealing point determination in this study is the ASTM D 1177-65. The congealing point is determined in the same way as the freezing point, but cooling is continued as long as the sample has undergone so-called thickening, i.e. the viscosity has increased so much that the solution cannot be pumped around in a system any more. Therefore the congealing point is the temperature below which the fluid cannot perform as a heat transfer fluid any longer. For low-concentration betaine-water solutions, the congealing point is about 4 ºC lower than the freezing point, for high-concentration solutions it is about 10 ºC lower.

With betaine-water solutions the equipment burst protection reaches well beyond the freezing and congealing points of the fluid. No permanent physical changes could be induced in piping or other heat transfer equipment. Typically burst protection was tested 20-30 ºC below freezing point.

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The water solubility of betaine is 160g / 100g water. This gives the maximum betaine concentration of 61.5 % by weight in room temperature. Because the solubility decreases slightly in lower temperatures a concentration of 50 to 55 % by weight is considered the practical limit for actual heat transfer system use.

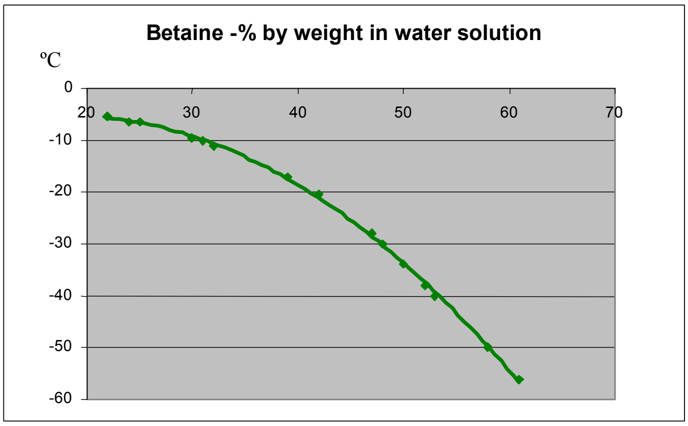


Figure 2: The freezing point in ºC of a betaine-water solution by betaine concentration.

## 1.2 Specific Heat Capacity

The specific heat capacity of a heat transfer fluid is one property affecting performance. Usually heat capacity values of fluids are compared to the value of pure water (app. 4.19 kJ/kgK). Most heat transfer fluids are water based and have a lower heat capacity than pure water.

The heat capacity of betaine-water solutions was found to be in the same range as that of traditional fluids, although slightly lower than glycols'. In Figure 3, the comparison is illustrated with heat transfer fluids having the same freezing point of -15 ºC. The fluids used for comparison are the most commonly used traditional fluids ethylene glycol and propylene glycol based water mixtures.

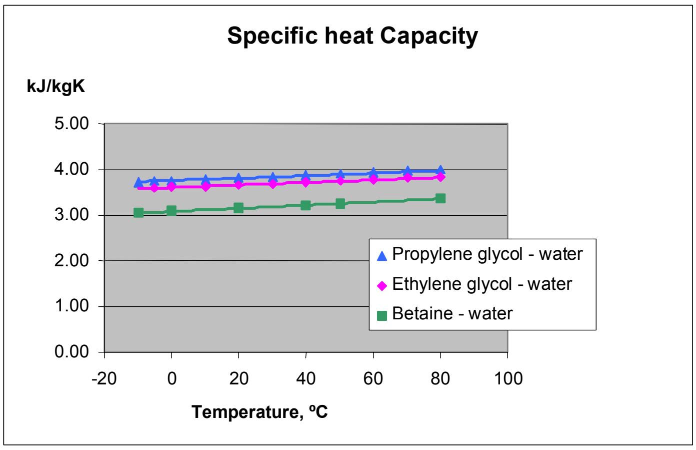


Figure 3: The specific heat capacity of a betaine-water solution compared to traditional fluids.

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1.3 Kinematic Viscosity

Viscosity is also one of the key elements in determining the technical performance of a heat transfer fluid. Typically viscosity values rise when temperature drops and therefore forced circulation of the fluid becomes more energy consuming and heat transfer less efficient.

In Figure 4, the comparison is illustrated with heat transfer fluids having the same freezing point of -15 ºC. Properties of ethylene glycol and betaine based fluids can be seen as very similar. Propylene glycol based fluids differ from the group with much higher viscosity.

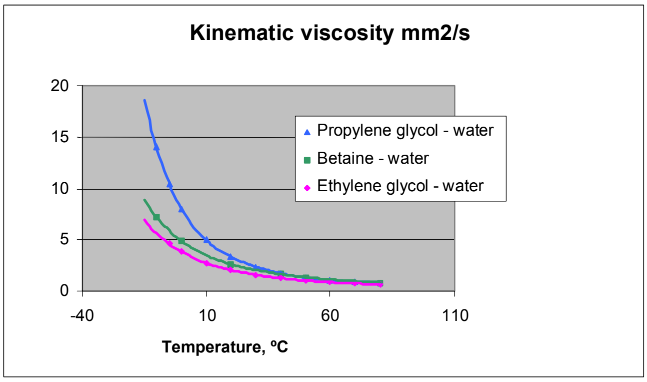


Figure 4: The viscosity of a betaine-water solution compared to traditional fluids.

1.4 Heat Conductivity

Heat conductivity is an important property, but very hard to measure in liquids. The results given in figure 5 are measured with fluids having the same freezing point of -15 ºC.

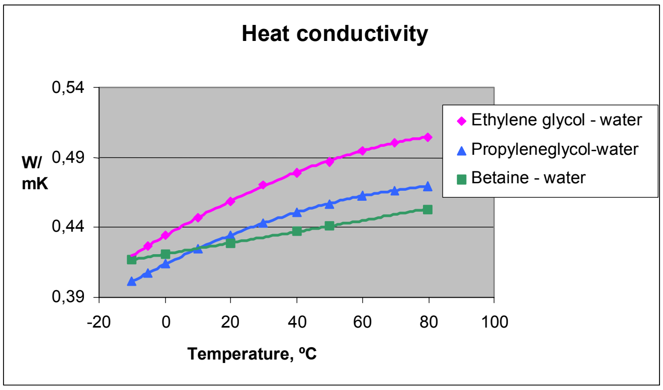


Figure 5: The heat conductivity of a betaine-water solution compared to traditional fluids.

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1.5 Conclusions from Physical Properties Measurements
All the measured properties above offer some insight into the fluids actual performance in heat transfer systems. However, it is important to notice that the importance of each property can vary from application to application and can sometimes be even misleading. The operating temperature range and flow characteristics in the particular system have to be taken into account. In figure 6 the Reynolds number is given for betaine- and traditional fluids in a certain flow environment.

The Reynolds number defines the quality of flow (from laminar, low figure to turbulent, high figure). Flow in the system should stay turbulent if high and efficient heat transfer is desired. It can be seen from figure 6 that a betaine solution has very good flow properties even in low temperatures. Flow dynamics is a factor often ignored by equipment dimensioning software.

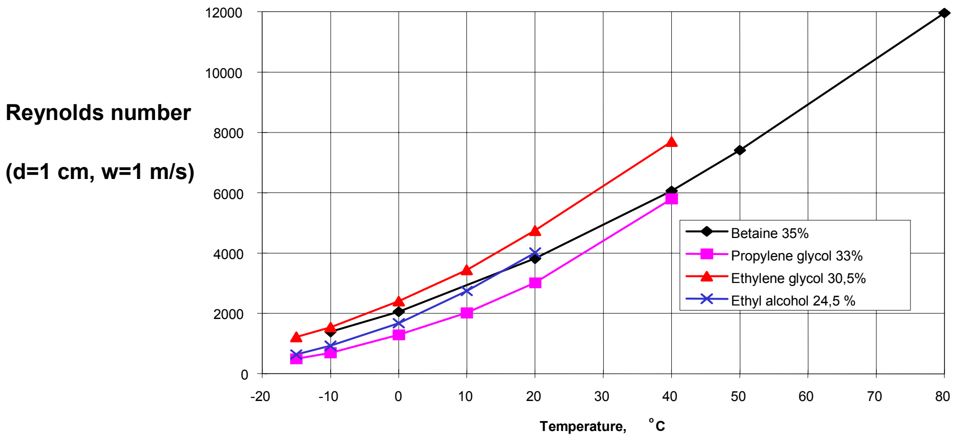


Figure 6: The Reynolds number of a betaine-water solution compared to traditional fluids (11).

Heat transfer efficiency of betaine based fluids has in field-tests been observed to even top that of most commonly used ethylene glycol based fluids. Below is presented a typical test result from an existing office building heat recovery system (Table 1). Thermal efficiency given in the table was measured by the following procedure:

1. Existing fluid was examined and found to be ethylene glycol with freezing point of -30 ºC
2. System thermal efficiency data was gathered by measuring inlet and outlet temperatures of air and fluid for a long period
3. System was drained and a betaine-water fluid with freezing point of -30 ºC was filled in. No operating parameters were changed (same flow etc)
4. System thermal efficiency data was again gathered by measuring inlet and outlet temperatures of air and fluid for a long period
5. From the gathered data a period with similar outdoor climatic conditions was selected and thermal efficiency of heat transfer from outlet air into inlet air was calculated. Results in table 1.

Table 1. System efficiency field-test. Existing office building heat recovery system.

| Circulating fluid | Measured Thermal System efficiency |
|---|---|
| Ethylene glycol-water mixture, -30 °C freezing protection | 28,8 % |
| Betaine water solution, -30 °C freezing protection | 30,1 % |

The importance of flow characteristics is evident from the results of table 1. Here, the results would be expected to be favorable for ethylene glycol rather than betaine based fluid if only the physical properties listed in 1.1 - 1.4 are taken into account. In this particular comparison the operating main temperature of the system is so low that the flow dynamics seem to take precedence over heat capacity properties. This is not a general physical fact, but something that follows from the difference in the physical properties of the compared fluids.

## 2. ENVIRONMENT AND SAFETY RESEARCH

Safety of use and operation, as well as environmental aspects are increasingly important in heat transfer fluid quality evaluation. Betaine as a natural substance has been found to have excellent qualities in this respect. Most important results will be discussed in the following chapter.

### 2.1 Toxicity

Here the main toxicity measurements and studies are summarized. Most are based on unpublished studies made during 1988-1992 for Cultor Ltd. Finnsugar Bioproducts, the betaine producer for this study.

Betaine is present in many organisms, including human body. Especially high levels can be found in several plants, some microbes and marine invertebrate animals. Of the betaine accumulators the best known ones are plants belonging to the Chenopodiaceae family (e.g. sugar beet). Betaine content of sugar beet is in average about 1.9 g/kg(four-year average in October in Germany) (1). In crustaceans, normally used for human consumption,the measured betaine concentrations of fresh muscle have varied from 1 to 9 g/kg and those in molluscs from 6 to 14 g/kg (12).
Betaine concentration in the muscle of young salmon, fed with control diet and betaine containing diet (1.5 %) for 1 - 2 months, was 0.12 - 0.28 and 0.7 - 0.9 g/kg, respectively (13).

Metabolism of betaine is well known. It acts as a natural methyl group donor in several metabolic reactions, such as the synthesis of methionine from homocysteine. Methyl donors generally induce the mobilization of liver lipids. Betaine is thus widely distributed in different organs and cells, the highest concentrations being found in renal medullas, where betaine acts as an osmolyte. In the methylation reaction, betaine is converted to dimethyl glycine and this is further metabolized to sarcosine and glycine. The absorption rate from intestine is unknown, but the apparent digestibility is close to 100 % (14).

Betaine has been widely used for years in commercial feeds for several animal species, including poultry, pigs, calves and fish, at concentrations from 0.2 to 15 kg/tn feed. In the feed betaine is acting as a natural methyl group donor thus enabling the partial replacement of choline and methionine in the feed. Another important function of betaine is acting as an osmoprotectant having stress relieving properties under various kinds of gastrointestinal stress.

Moreover, betaine stabilizes the macromolecular functions, by increasing the temperature and ionic tolerance of enzymes and membranes. Betaine supplementation has been widely adopted in fish feeding to relieve the osmotic stress of fish at seawater transfer. No harmful effects have been observed in several long-term trials.

Betaine is widely used in human foods e.g. in Japan. Betaine has also been used in some pharmaceutical applications, e.g.. in the treatment of human homocystinuria for several months without any harmful effects (2) and in preparations to prevent the development of fatty liver. Betaine has food approval in Japan and Korea and Dietary supplement status by FDA in the US.

Betaine is used globally in cosmetic products, like hair and skin care products.

Betaine has successfully been used in fermentations producing e.g. vitamins, antibiotics and amino acids because of its capability to protect microbes from osmotic stress and its good compatibility with enzymes.

- For betaine anhydrous, acute oral median lethal dosage, LD50 (oral) rats, is 11.179 + 0.725 g/kg B.W. Therefore the product is not classified as toxic for acute health hazards (3).
- Bacterial reverse mutation assay (Ames test with Salmonella typhimurium) showed that betaine monohydrate is not mutagenic when assayed up to 5000 mg/plate (4).
- Metaphase analysis of human lymphocytes showed that betaine monohydrate is not a clastogen to human lymphocytes (5).
- Sensitization test in the guinea pig showed that there is no evidence to suggest that betaine monohydrate acts as a sensitizer. Betaine would be classified as a non-allergenic or as a weak, grade 1 sensitizer on the Magnusson and Kligman scale, having produced a sensitization rate of 0 % (6).
- Acute eye irritation study with 10 % by weight betaine monohydrate in distilled water did not result in any ocular irritation, and it can be considered non-irritant at this concentration (7).
- The primary skin irritation potential test showed that betaine monohydrate at levels over 3.5 % produced a highly significant reduction in the irritant potential of solutions containing 5.15 % to 10.3 % Sodium Lauryl Ether Sulphate (SLES). The extent of this effect was shown to be concentration dependent. In addition, 50 % (w/v) solution of betaine monohydrate appeared to give rise to even less irritation than the deionized water control sample (8).
- The Minimum Inhibitory Concentration (MIC) of betaine monohydrate against nine selected microorganisms was shown to be greater that 10 % w/v. Betaine monohydrate did not appear to affect the growth rate of any of the micro organisms at concentrations up to 10% w/v (9).
- The effect of exaggerated treatment with a cosmetic lotion containing betaine monohydrate on the rate of transepidermal water loss (TEWL) of intact skin identical treatment with the placebo lotion was tested.

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There were clear indications that the addition of betaine to a skin care lotion resulted in improved moisture retention when the product was applied to the skin(15).
- Acute fish toxicity was analysed to be: LC50 96h Brachydanio rerio > 100 mg/l. (18)
- Acute Daphnia toxicity was analysed to be: EC50 48h Daphnia magna > 100 mg/l. (18)
- Betaine has food approval in Japan and Korea and Dietary supplement status by FDA in the US.

2.2 Biodegradability and Oxygen Demand
Betaine is naturally occurring in many animals, plants and microbes. When betaine is released into the environment either by death of the organism or excretion, it becomes available as a substrate for microbial growth.

The ability to decompose betaine is widespread among microorganisms and both anaerobic and aerobic are known to degrade betaine. Laboratory tests by Cultor Ltd. Finnsugar Bioproducts have shown that betaine degrades fully within two weeks in sandy clay soil due to the action of soil microbes. Mineralisation of betaine after 28 days was over 99,6 %, therefore betaine can be considered easily biodegradable (16).

Betaine is rich in several marine invertebrate animals and, being as prey animals of fish, they form an important source of betaine for the fish. In artificial feeds for salmonids, betaine (+ some amino acids) is commonly added at 2 - 15 g/kg concentrations with beneficial effects on feed palatability, growth and feed conversion rate. Thus, assuming a feeding rate of 1 - 8 %/ body weight/ day and a betaine content of 2 - 15 g/ kg feed, the betaine uptake by fish via food varies from 20 to 1200 mg/ kg/ day, and no harmful effects have been observed in several long-term (up to one year) trials.

Betaine concentration in the muscle of young salmon, fed with control diet and betaine containing diet (15 g/kg feed) for 1 - 2 months, was 0.12 - 0.28 and 0.7 - 0.9 g/ kg, respectively(13). For comparison, in crustaceans, normally used for human consumption, the measured betaine concentrations of fresh muscle have varied from 1 to 9 g/kg and those in molluscs from 6 to 14 g/kg (17).

Oxygen demand (19):

| | Unit | Thermera -15 | Thermera -35 |
|---|---|---|---|
| COD(Cr) | mg/g | 15 | 16.5 |
| BOD5ATU | mg/g | 422 | 558 |
| BOD7ATU | mg/g | 431 | 540 |
| BOD28ATU | mg/g | 478 | 759 |

The solubility of betaine is 1600g per kg pure water, thus the partition coefficient of betaine in water/n-octanol system was analysed to be very low:
Pow = 8.1 x 10-4
log = -3.1
Therefore betaine has the potential to be accumulated in biological systems. However the relevance of betaine in biologic systems is shown above and therefore the potential to be accumulated in biological systems should have no significance.

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3. CORROSION AND ADDITIVES NEEDS

Corrosion rates for various materials with betaine based solutions were measured. The results show that corrosion rate even without any corrosion inhibitors added is well below levels accepted by equipment manufacturers in the HVAC industry (for copper for example, a corrosion rate up to 2.0 µm p.a is most commonly accepted). Corrosion speed for pure water was higher.

Table 2. Corrosion rates for a 50% betaine-water solution. The higher figure indicates the corrosion rate at the beginning of the tests and the lower figure the rate stabilised with time (~48h) (16).

| **Material** | **Loss in material thickness, µm p.a** |
|---|---|
| Copper | 1.5 to 0.5 |
| Carbon steel Fe52 | 75 to 10 |
| Brass | 1.5 to 0.2 |
| Red bronze | 125 to 2 |
| Cast iron | 0.9 to 0.2 |

Further tests were done according to demands of other than the HVAC industry. In the automotive industry, for example, the demands are much higher and standardized test methods are used. Very common is the ASTM 1384 test. Betaine fluid was tested with a conventional film forming corrosion inhibitor according to this test.
Concentrations of the inhibitor package's active ingredients were from 0,05% to 0,1%. Results are given in Table 3 as loss in material thickness and mass loss.

Table 3. ASTM 1384 corrosion test results for a 35% betaine-water solution containing conventional film forming corrosion inhibitor used in the HVAC industry.

| **Material** | **Loss in material thickness, µm p.a** | **Mass loss, mg** |
|---|---|---|
| Copper | 0.3 | 0.2 |
| Fe37 | 0.3 | 0.2 |
| Solder | 0.3 | 0.2 |
| Bronze | 0.3 | 0.2 |
| Cast iron | 22 | 16.5 |
| Aluminum | 2.4 | 1.8 |
| Zn | 4.0 | 3.0 |

Further standardized test performed included the Double Chamber Cavitation Corrosion Test (CEC C-23-T-99), the Hot Plate Corrosion Test (ASTM D 4340) and the Simulated Service Corrosion

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Test (ASTM D2570-96). The betaine based fluid fulfilled requirements set by the AFNOR NFR 15-601 norm for automotive coolant use.

3.1 Conclusions from corrosion and additives testing
The requirements of different industries were closely examined. Heat transfer fluids are used in a variety of systems and conditions. Most important variable is the temperature range of operation. The fluid needs to be thermally stable and have acceptable corrosion properties.

It was found that generally the need for additives is very low for betaine based fluids. Additives are most commonly used in conventional fluids for corrosion protection, pH stability, anti-foaming, coloring and flavoring. Betaine based fluids performed so well in the testing that in many application areas no additives were found necessary to fulfil requirements (especially in the HVAC industry applications such as: air conditioning, heat recovery, heat pumps, phase change heat transfer systems). Only to fulfil the harshest test requirements of the automotive industry, corrosion inhibitors were required. However, the needed concentration of inhibitors was substantially lower than is common with conventional fluids.

## 4. LIFE CYCLE COST

When selecting a heat transfer fluid, next to the thermodynamic properties, toxicity requirements and environmental considerations, the total cost of ownership needs to be taken as selection criteria. The life cycle of a HVAC system consists of 5 life cycle phases:

- Design phase
- Construction phase
- Commissioning and start-up phase
- Operational phase
- Dismantling phase

In each of these phases comprehensive decisions need to be taken that have an effect on the total cost of ownership of the installation

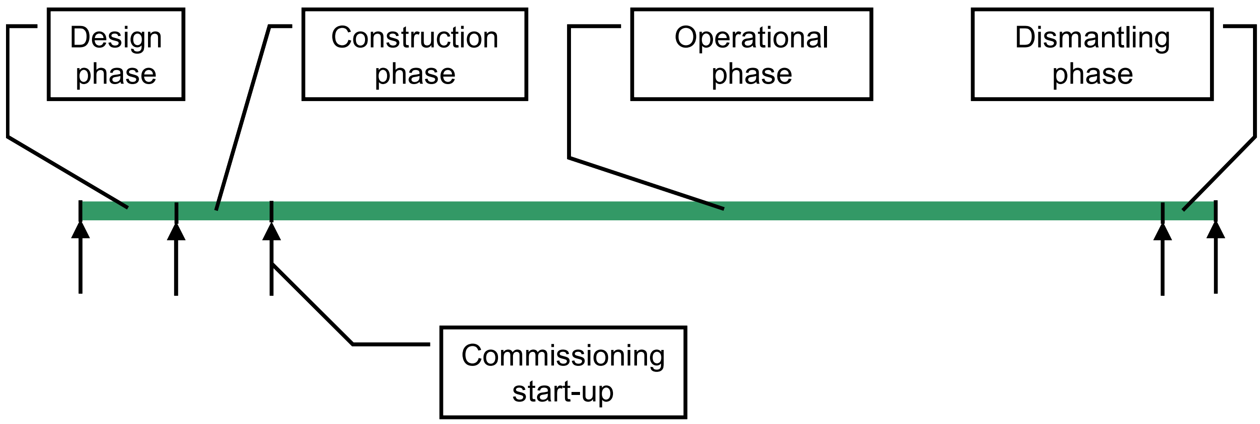


Figure 7: life cycle of an HVAC installation

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Design phase:
This is the most important and complex phase in the life cycle of a heat transfer fluid in a HVAC system, where the total cost of ownership are generated. Decisions must be taken to adapt the collaboration of all chosen components into the calculation of the total cost of ownership:

- Choice of the heat transfer fluid, taking the following parameters into consideration
    - Thermodynamic properties
    - Toxicity requirements
    - Ecological requirements
    - Initial cost of the heat transfer fluid
    - Environmental or toxicity subsidies connected to the choice of the heat transfer fluid
    - ISO 14000 gains connected to the choice of the heat transfer fluid
    - Filling and commissioning cost of the heat transfer fluid
    - Component cost savings and cost adders for the chosen heat transfer fluid (heat exchanger, pump, components and piping sizing)
    - Cost of operation
        - Energy consumption
        - Maintenance costs
            - Inhibitor maintenance
            - Concentration changes
            - Life time of the heat transfer fluid (chemical and biological stability)
    - Risks and cost of installation failures
        - Direct cost required to prevent installation leakages to environmental exposure
        - Environmental risks and cost during leakages
        - Toxicity risks and cost during leakages
        - Concentration losses when maintenance is postponed
        - Corrosion risks when maintenance is postponed
        - Burst risks during unforeseen installation freezing
    - Cost of Dismantling
        - Environmental cost of destruction of the heat transfer fluid itself
            - Cost of transportation (toxic waste or not)
            - Cost of destruction (destruction compared to flushing into the sewer)
        - Environmental cost of destruction of contaminated installation components
        - Environmental cost of cleaning in case leakages have occurred during the operational phase of the installation (building and/or soil).

An indicative life cycle cost comparison between the various conventional glycol products and a Betaine based fluid were made below. The calculation is based on the prices of the products and the disposal costs in Finland. In the real-life example shown in the picture, a glycol product is about 24% more expensive. In this comparison the operational phase of the installation was set to10 years. After the operational phase, the dismantling phase cost are included into the graph. Glycols need to be treated as chemical waste, whereas Betaine based fluids can be drained into the sewer system, without additional cost. In small-scale systems, in which the disposal costs of the spent fluid will grow, the difference may be as high as 77%.

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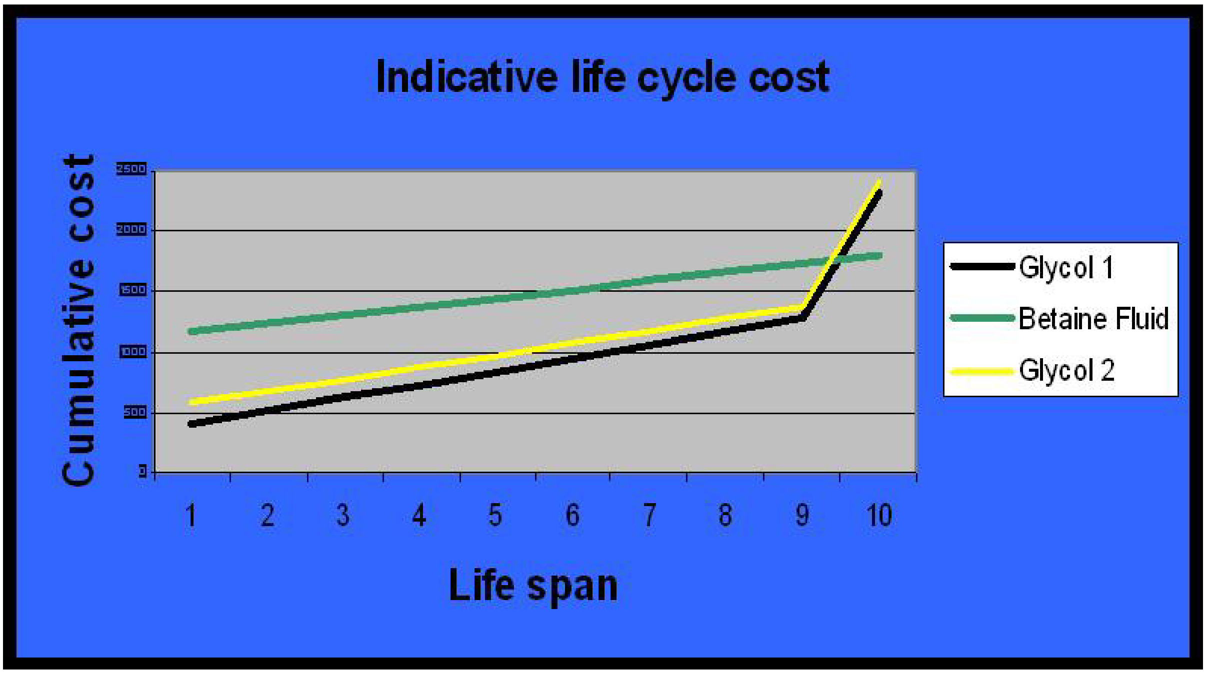


Figure 8: Indicative life cycle cost comparison between conventional glycol products and Betaine

CONCLUSIONS

The advantages of Betaine based heat transfer fluids, compared to glycols, are non-toxicity and environmental friendliness, combined with natural corrosive properties.

The physical properties and technical performance of betaine based fluids were found to be very acceptable according to the industry standards found. In actual system design it is important to understand that a complex mix of different physical properties determines the total performance of a fluid. The software used should have all these data and not use traditional assumptions based on properties of conventional fluids. Technical performance of betaine based fluids was found to be higher in empirical tests than in simulations with conventional tools.

The environmental and health qualities of betaine are certainly superior to that of most often used conventional fluids. Even in the applications where a small package of corrosion inhibitors is required, a betaine-based fluid can still be held non-toxic and safe to the environment.

Betaine has a natural anti-corrosive nature. In closed systems with temperatures typical for the HVAC industry in most cases no corrosion inhibitors are required. In areas with stricter requirements, such as the automotive industry, corrosion is easy to handle with mild dosages of common inhibitors.

The life cycle costs of Betaine based fluids are competitive with conventional glycol products if the cost of ownership are taken in consideration in advance and the installation is properly designed for the use of Betain based fluids.

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