Evaluation of Heat Pipe Working Fluids In The Temperature Range 450 to 700 K

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Abstract. In the temperature range of 450-700 K, there are currently no working fluids that have been validated for heat pipes and loop heat pipes, with the exception of water in the lower portion of the range. This paper reviews a number of potential working fluid including several organic fluids, mercury, sulfur/iodine, and halides. Physical property data are used where available, and estimated where unavailable using standard methods. The halide salts appear to possess attractive properties, with good liquid transport factors, and suitable vapor pressures. Where nuclear radiation is not a consideration, other potential working fluids are aniline, naphthalene, toluene, and phenol. The limited available life test data available suggests that toluene, naphthalene, and some of the halides are compatible with stainless steel, while the other fluids have not been tested.

HEAT PIPE/LOOP HEAT PIPE WORKING FLUIDS

The U.S. Department of Energy (DOE) and the National Aeronautics and Space Administration (NASA) are examining space nuclear electric power systems for the exploration of deep space. Some of the proposed exploratory designs would use heat pipes (HPs) or loop heat pipes (LHPs) as components in a radiator to reject the waste heat in the temperature range of 450 K to 700 K. Another application of heat pipes or LHPs in this temperature range is the cooling of SiC electronics. The benefits of SiC over silicon include its wide band gap energy, high breakdown electric field, and high thermal conductivity.

In the intermediate temperature range, from roughly 450 K to 700 K, there are currently no proven working fluids that can be used in heat pipes and LHPs, with the partial exception of water. Water has been used at temperature up to about 550 K. However, the vapor pressure of water is 26 atmospheres at 500 K, and rises rapidly with further increase in temperature. As the vapor pressure increases, the required envelope thickness to withstand the pressure increases. The resulting increase in mass can limit the practical upper operating temperature of water. In addition, some of the envelope materials that are compatible with water at the lower temperatures become incompatible at higher temperatures.

Similarly, the alkali metals such as cesium, potassium, and sodium are suitable working fluids at temperatures above this temperature range. Heat pipes with various alkali metal working fluids have been operated at temperatures above 700 K, and a cesium LHP has been tested at 850 K (Anderson, 1995b). However, as the temperature is decreased below 700 K, the sonic limit prevents these fluids from operating in this temperature range. As the temperature is reduced, the vapor pressure and vapor density are decreased. At low enough temperatures, the vapor density is so low that the vapor sonic velocity limits the heat transfer. The heat pipe (or LHP) vapor space becomes too large to be practical for alkali metals in the intermediate temperature range.

A number of previous studies have utilized various working fluids. Deverall (1970) reported work with mercury, while Polasek and Stulc (1976) examined sulfur. Rosenfeld and Lindemuth (1992) examined sulfur, sulfur/iodine,

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TABLE 1. Intermediate Temperature Fluids**.**

Fluid		Melting Point, K	Boiling Temp., K	Critical Temp., K	Critical Pressure, MPa
Water	H_2O	273	373	647	22.12
Dowtherm A	Diphenyl/Diphenyl Oxide	285	530	770	3.135
Sulfur	S	386	718	1314	20.7
Sulfur/10% Iodine	S/10%I	-390	—	—	—
Iodine	I	387	458	785	11.6
Naphthalene	$C_{10}H_8$	354	491	748	4.05
Phenol	C_6H_6O	314	455	694	6.13
Toluene	$C_6H_5CH_3$	178	384	592	4.1
Hydrazine	N_2H_4	275	387	653	14.7
Aniline	C_6H_7N	267	458	699	5.3
Titanium Tetrachloride	$TiCl_4$	298	410	638	4.7
Titanium Tetrabromide	$TiBr_4$	312	503	796	—
Titanium Tetraiodide	TiI_4	423	650	1040	—

and iodine. Saaski and Tower (1977) suggested naphthalene, toluene and the halide salts, $SnCl_4$, $TiCl_4$, and $SbCl_4$ as potential working fluids. Anderson and Bland (1995) considered additional halide salts as well as phenol.

As shown in Tables 1 and 2, the present review examines all of the fluids above, as well as several additional candidate fluids. Unfortunately, fluid property and life test data are incomplete for all of the potential working fluids. To screen the fluids, Merit Number (also known in the literature as Liquid Transport Factor) and vapor pressure as a function of temperature were calculated. Unavailable physical property data were estimated, as discussed below, using the methods of Reid et al. (1987). While this estimated data set is not suitable for heat pipe/LHP design, it is useful in identifying potential working fluids.

VAPOR PRESSURE AND MERIT NUMBER

Vapor pressure and merit number (or Liquid Transport Factor) as a function of temperature are two properties used to screen potential working fluids. The merit number is a means of ranking heat pipe fluids, with higher merit number more desirable:

$$M = \frac{\rho_L \cdot \sigma \cdot \lambda}{\mu_L} \qquad (1)$$

Vapor pressure for potential working fluids as a function of temperature is shown in Figures 1 through 4. Note that the vapor pressure for water is too high (for much of this range), and the vapor pressure for cesium is too low, so a vapor pressure intermediate between the two is desirable. It can be seen that most of the fluids listed above have a suitable vapor pressure.

Figures 5 and 6 shows the merit number as a function of temperature. While water and cesium have good merit numbers, their vapor pressures eliminate them from consideration. The fluids are discussed in more detail below.

Mercury

Mercury was considered and rejected as a working fluid, due to wetting problems. For any capillary pumped system to operate, the working fluid must wet the wick, so that capillary forces can return the liquid to the evaporator. Mercury does not wet stainless steel and most other potential envelope/wick materials. Deverall (1970) reported

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TABLE 2. Halide Properties**.**

	Melting Point, K	**Boiling Point, K**	**Critical Point, K**	**Latent Heat (Boiling Point) kJ/kg**	**Density, kg/m^3**	**Surface Tension, N/m**	**Viscosity, cPoise**
$AlBr_3$	370	528	763	85.5	2331 (500 K)	—	0.809 (500 K)
AlI_3	464	658	983	158.2	3133 (500 K)	—	2.10 (500 K)
BCl_3	165.9	285.8	455	—	—	—	—
BBr_3	227	364	581	—	—	—	—
BI_3	323.1	483	773	—	—	—	—
$BiCl_3$	503	714	—	229.6	3693 (600 K)	0.0586 (600 K)	19.4 (600 K)
$BiBr_3$	491	734	—	168	4477 (600 K)	0.0586 (600 K)	—
$GaCl_3$	351	575	—	356.7	1743 (500 K)	0.0124 (500 K)	0.449 (500 K)
$GaBr_3$	395	587	—	189.4	2770 (500 K)	0.0195 (500 K)	1.027 (500 K)
$SbCl_3$	346	492	—	190.7	2329 (500 K)	—	0.337 (500 K)
$SbBr_3$	370	553	—	163.2	3193 (500 K)	—	0.957 (500 K)
SbI_3	443	674	—	136.7	3776 (600 K)	—	—
$SiCl_4$	204.3	330.8	508.1	170.1	1259 (400 K)	—	—
SiI_4	393.7	560.5	944	93.7	3589 (500 K)	—	—
$SnCl_4$	240	388	—	130.1	1944 (400 K)	0.0155 (400 K)	0.360 (400 K)
$SnBr_4$	303	478	—	93.5	—	—	—
SnI_4	418	621	—	80.15	—	—	—
$TiCl_4$	243	409.6	638	190.9	1543 (400 K)	0.0211 (400 K)	0.391 (400 K)
$TiBr_4$	312	506	795.7	120.8	2758 (400 K)	—	—
TiI_4	427	650	1040	101	3258 (500 K)	—	—

successfully operating mercury heat pipes with magnesium added to promote mercury's wetting. However, these heat pipes used coarse screen wicks, which are more easily wetted. More recently, Rosenfeld (1994) fabricated a sintered wick heat pipe with mercury as the working fluid. The mercury did not wet the small pores in the wick, even though wetting agents were added. The pore size in this sintered wick was more than 10 times larger than the pore size in a loop heat pipe. It is doubtful that a loop heat pipe or high performance heat pipe wick could be treated so that it would be wet by mercury. Apart from these thermo-mechanical considerations, mercury is toxic and a difficult material to handle in processing and cleaning. Hence, for the current purposes, mercury was considered and rejected as a working fluid.

Sulfur/Iodine

Sulfur has a unique temperature dependent polymerization property at 475 K, which increases its liquid viscosity peak to approximately 100 Pa-s. This is about three orders of magnitude higher than the maximum level for effective heat pipe operation. Even at 575 K, the viscosity is still an order of magnitude too high for useful heat pipe operation. The addition of 3-10% of iodine will reduce the viscosity of sulfur by more than three orders of magnitude to a level sufficient for effective heat pipe operation (Polasek, 1976). Sulfur/iodine heat pipes have been built and tested; however, pure iodine is more effective than sulfur or sulfur-iodine for heat pipe operation (Rosenfeld, 1992).

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There are some disadvantages to iodine. It is a corrosive fluid with unknown long-term reliability. In addition, iodine has a low thermal conductivity, increasing the temperature drop in the heat pipe.

Organic Fluids

The organic fluids that were examined are aniline, naphthalene, toluene, hydrazine, and phenol. One disadvantage is that these fluids are more likely to generate non-condensable gas when exposed to radiation in contrast to the halides and other inorganic working fluids. Hence organic fluids are not considered to be suitable for space nuclear electric propulsion applications. However, they are potentially useful in developing heat pipes for thermal management of electronic components made of SiC.

Vapor pressures and Merit Numbers for the organic fluids are shown in Figures 1 and 5, respectively. Toluene and hydrazine have vapor pressures that are only a factor of 2 or 3 lower than water at a given temperature. This, combined with a Merit Number more than 10 times lower than water, limits their potential usefulness. Phenol, Aniline, and Naphthalene have vapor pressures more than 10 times lower than water. As shown in Figure 5, their Merit Numbers are higher than most of the other candidate fluids.

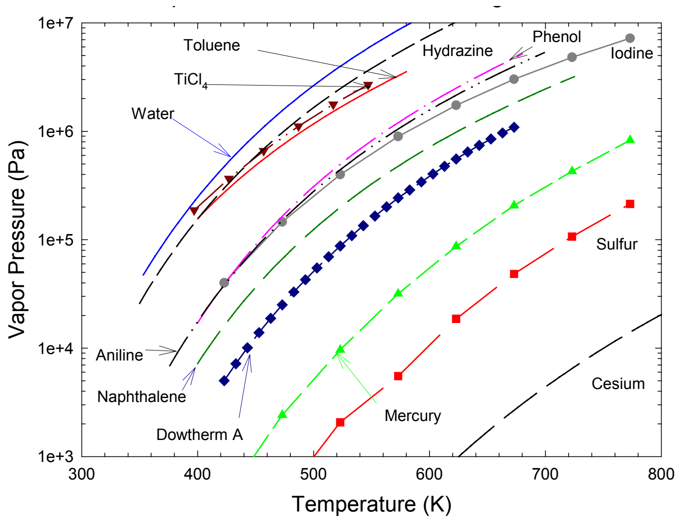

FIGURE 1. Vapor Pressure as a Function of Temperature, Potential Heat Pipe or LHP Working Fluids.

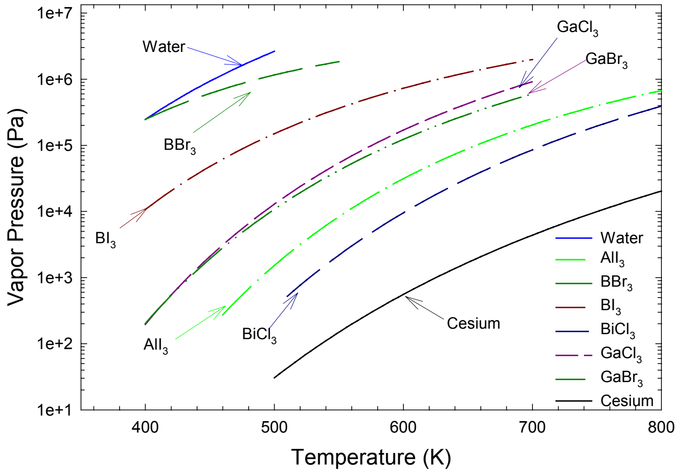

FIGURE 2. Vapor Pressures - Al, B, Bi, and Ga Halides (Brandes, 1983).

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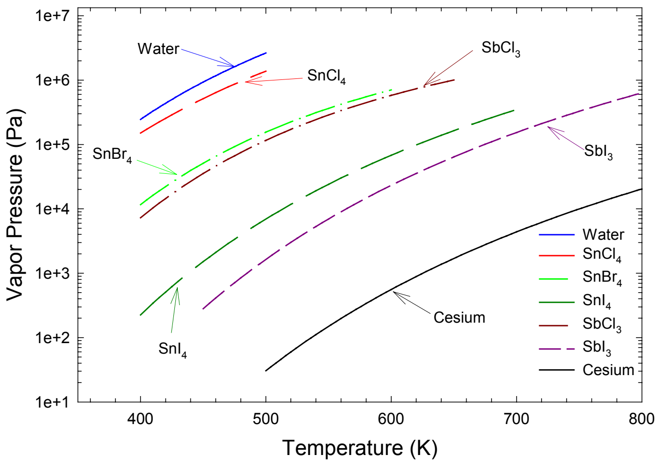

FIGURE 3. Vapor Pressures - Sb and Sn Halides (Brandes, 1983).

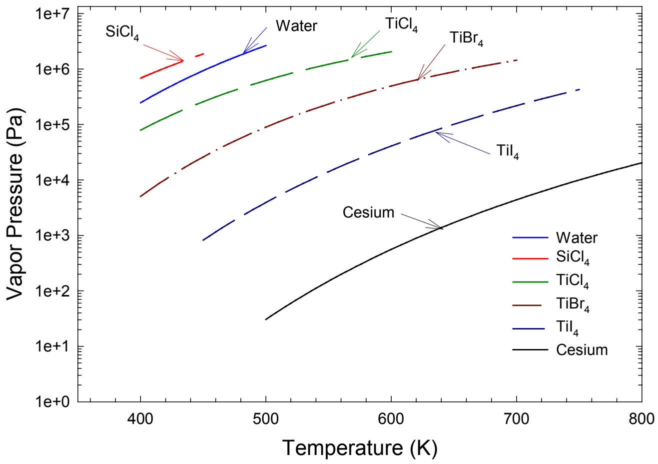

FIGURE 4. Vapor Pressure as a Function of Temperature, Si and Ti Halides (Brandes, 1983).

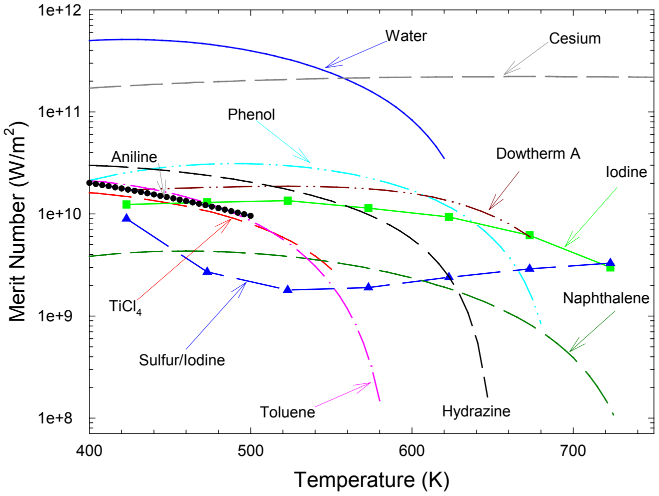

FIGURE 5. Merit Number as a Function of Temperature, Potential Working fluids.

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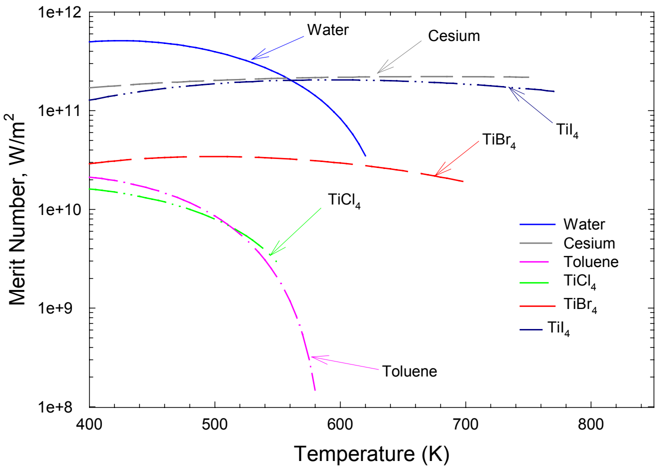

FIGURE 6. Merit Number as a Function of Temperature, Potential Halide Working Fluids. Note That Some of the Properties are Estimated.

Halides

Another set of potential working fluids is the halide salts of titanium, aluminum, boron, antimony, tin, and silicon. Table 2 contains physical property data for many of the halides. It is based on data (or correlations) taken from Brandes (1983), Janz (1988), and Reid *et al*. (1987). Unfortunately, the physical property data are incomplete. Better physical property data are required before heat pipes using these fluids can be reliably developed. Vapor pressures of halides are shown in Figures 2 to 4, based on correlations in Brandes (1983). Almost all of the fluids are in the desirable range between water and cesium. In addition, while one fluid may not be suitable for the entire range, a family of similar fluids could be. If one fluid is compatible with a given envelope, the other fluids are probably also compatible. For example, Figure 4 shows that the titanium halides cover the entire temperature range.

Russian heat pipe developers have reported success with titanium tetrachloride ($TiCl_4$) (Chechetkin, 1971), but a thorough examination of related compounds apparently has not been undertaken. It is possible that some of the related salts such as $TiCl_2F_2$ may have better working properties. These working fluids are more polar, which increases the surface tension and the liquid transport factor.

Figure 6 indicates that the Titanium halides are good candidate fluids. It is noted that the merit number for $TiCl_4$ is based on data. On the other hand, the surface tensions and viscosities used in the merit number calculations for $TiBr_4$ and TiI_4 are estimates based on $TiCl_4$, using the methods in Reid *et al.* (1987). The high Merit numbers indicate that additional fluid property data should be acquired to examine these fluids in more detail.

COMPATIBILITY STUDIES AND LIFE TESTS

None of the potential working fluids has adequate life test data at appropriate conditions. Life tests are required to verify that the heat pipe envelope, wick, and working fluid are compatible for the potentially long operating life of a heat pipe. Deep space missions, for example, take about fifteen years. The two major consequences of incompatibility are corrosion and the generation of non-condensable gas, or both. The resulting corrosion products can block portions of the wick, preventing the heat pipe from operating properly. In more extreme cases, the heat pipe can leak.

To conduct a life test, an envelope material, a wick material, and a working fluid are chosen. A simple heat pipe is then fabricated using the chosen material, and tested as the desired operating temperature. Temperatures are monitored to detect the formation of non-condensable gas. Testing generally continues until the heat pipe either fails, or the duration of the life test is complete. In either case, the heat pipe is then sectioned and examined for possible incompatibilities.

TABLE 3. Sulfur/Iodine Heat Pipe Life Test Data (Rosenfeld, 1992)

Material	Operating Temperature, K	Operating Power, W	Duration, Hours	Condition
Aluminum 5052	623	30	1,028	Grain boundary penetration
Ti-6Al-4V	623	30	1,000	150 K ΔT
Titanium C/P-2	523	20	24	Failed
SST 304	623	30	1,008	No sign of failure
Niobium-1% Zr	623	40	950	Failed

TABLE 4. Life Test Data for Intermediate Temperature Fluids (Saaski, 1977)

Working Fluid	Envelope	Operating Time	Operating Temp., K	Power, W	$\Delta T_{Initial}/\Delta T_{Final}$, K
Toluene	Aluminum	2014	403	26.2	5.06/5.74
Toluene	Carbon Steel	672	392	26	5.08/6.42
$SnCl_4$	Aluminum	----	432		Incompatible
$SnCl_4$	Carbon Steel	2014	432	26	2.78/13.92
$TiCl_4$	Aluminum	664	438	29.6	10.3/14.32
$TiCl_4$	Carbon Steel	2014	425	38.8	4.3/5.93
Naphthalene	Aluminum	2014	491	52.5	15.2/15.8
Naphthalene	Carbon Steel	2014	490	65	6.38/6.13
$SbCl_3$	Aluminum	----	500		Incompatible
$SbCl_3$	Carbon Steel	1518	500	41.5	101.83/82.67

Previous life test data on fluids in this temperature range is scarce. There are a small amount of data on sulfur and iodine (Rosenfeld, 1992); see Table 3, and halides, toluene, and naphthalene (Saaski, 1977); see Table 4. The limited life test data available suggests that toluene, naphthalene, sulfur/iodine, and some of the halides are compatible with stainless steel. Most of the tests to date have been conducted with aluminum or steel envelopes. Due to its low mass and high strength, titanium should also be considered as a potential envelope and wick material.

Lindemuth and Rosenfeld (1993) conducted life tests on two Monel K-500 heat pipes with water at 473 K. One heat pipe was fabricated without a wick structure, and a second was fabricated using a nickel-plated copper felt metal wick structure. The second heat pipe employed a wick structure composed of low-phosphorus electroless nickel-plated OFE copper felt that was brazed to the inner wall. Both heat pipes used Monel K-500 end caps and Monel 400 fill tubes. Both devices were then life tested in a gravity-assisted orientation at a nominal temperature of 473 K. The wickless heat pipe developed a large temperature drop after 1000 hours of operation, and was vented. Subsequent life testing was performed for 14,000 hours with no additional signs of gas generation. The source of the early gas generation was not determined, but from the tests it was concluded that Monel K-500 was compatible with water at 473 K. The felt wick heat pipe operated with a favorable low temperature drop for 3500 hours, but exhibited signs of gas generation after 8300 hours. The source of the later generation was undetermined, but was suspected as originating from the low-phosphorus electroless nickel used to form the wick structure.

While we are not aware of life tests with water at higher temperatures, Lindemuth and Rosenfeld (1993) also fabricated and tested several Monel/water capillary-assisted inclined thermosiphons at temperatures up to 586 K. Each heat pipe had a length of 46 cm, an outer diameter of 1.6 cm inches, and a wall thickness of 0.035 inches. The wick structure was similar to that employed in the life test heat pipe. However, high-phosphorus electroless nickel plating was used to coat the copper felt metal wick because of concerns arising from the later gas generation observed in the wick-lined life test heat pipe. The end caps were brazed into the heat pipe tubes using 65%/35% Cu/Au braze. The heat pipes were operated as a capillary-assisted inclined thermosiphon, with a favorable 12-degree inclination. The heat pipes were each operated at 1300 W per heat pipe at 500 K with an evaporator heat flux of 34.2 W/cm^2. Tests were then conducted at 586 K with an evaporator heat flux of 52.5 W/cm^2 and a power transport of 2000 W per heat pipe. No performance limits were reached during the testing, which indicated that gravity-aided water heat pipes are capable of operating up to 586 K. Life tests, and tests with an adverse orientation would still need to be conducted.

CONCLUSIONS

Several of the halide salts, including Titanium Tetrachloride, Tetrabromide, and Tetraiodide appear to be suitable working fluids in the temperature range of 450 to 700 K. Other potential fluids include the Al, B, Bi, Ga, Sb, Si, and Sn halides. Related salts such as $TiCl_2F_2$ may have better working properties, since they are more polar, and should have a larger surface tension. Potential organic working fluids for non-nuclear applications include aniline, naphthalene, toluene, hydrazine, and phenol. These fluids are more likely to generate non-condensable gas when exposed to radiation.

Unfortunately, none of the potential working fluids can currently be used without further testing. The physical property data are incomplete. In addition, none of these fluids have sufficient life test data to allow confidence in their long-term operation. The limited life test data available suggest that toluene, naphthalene, and some of the halides are compatible with stainless steel, while the other fluids have not been tested. Due to its low mass and high strength, titanium should also be considered as a potential envelope and wick material.

NOMENCLATURE

M =Merit number (W/m^2)
ρ_L =Liquid density (kg/m^3)
σ =Surface tension (N/m)
λ =Latent heat (J/kg)
μ_L =Liquid viscosity (Pa)

DISCLAIMERS

The views and opinions expressed in this paper are of the authors and do not represent those of the U.S. Government or its agencies. In particular, no endorsement of any sort by any federal agency is implied.

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