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Radiative cooling with MgO and/or LiF layers

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Abstract

Selectively emitting surfaces for radiative cooling should have a high emissivity within the atmospheric window (wavelengths of 8 to 13 microns), and a low emissivity otherwise. These radiative properties permit the attainment of lower temperatures than blackbody radiators. Two materials which show promise for the fabrication of selective radiators are magnesium oxide and lithium fluoride. Appropriate infrared optical properties were obtained with a 1.1 mm thick MgO ceramic layer, polished on the topside and backed with a metal foil. In a simple passive cooling experiment, the MgO radiator reached 22°C below air temperature, 3°C colder than a highly emissive nonselective radiator.

The production of low temperatures by radiating surfaces exposed to the sky is currently limited by the less than ideal optical properties of available materials. Improvements could permit the future development of refrigeration systems which require no external power or roof systems for the air conditioning of buildings. The purpose of this report is to point out that layers of magnesium oxide and/or lithium fluoride, backed by an infrared reflective layer, can produce lower temperatures than other materials tested.

The usual approach to the fabrication of selectively emitting radiators is to employ an emitting layer with a high infrared absorptivity within the "atmospheric window" (8 to 13 μm wavelength) and absorptivity low elsewhere, on top of a reflective substrate.[1,2] The resulting radiator would then have the desired high emissivity between 8 and 13 microns and a low emissivity in the rest of the spectrum of thermal wavelengths (5 to 40 μm). This approach has the advantage that the absorbing layer can be thin: a few microns or less. However, it has the disadvantage that materials have not yet been identified which absorb in the correct waveband and nowhere else. Thus, for thin layers of available materials the absorption in the 8 to 13 μm region is not complete. The resulting radiator is selective, but not highly emissive and therefore cannot produce much cooling. If the layer is made thicker to produce complete absorption in the window region it begins to absorb outside the window, reducing the selectivity and limiting access to low temperatures.

Felix Trombe[3] has pointed out that single crystal MgO has favorable optical constants for radiative cooling. Lithium fluoride has quite similar infrared optical properties. Layers of these materials on the order of 1 mm in thickness, backed by a low emissivity layer such as a metal film, can produce good selective radiators due to the strong reststrahlen reflectance at wavelengths greater than 13 microns (14 microns for LiF). Figure 1 shows the computed spectral normal reflectance for layers of MgO and LiF with a reflective backing. Bulk reflectance values[4] were used for wavelengths greater than 10 microns and tabulated optical constants[5] were used to compute the reflectance at shorter wavelengths. The 50% reflectance point has been positioned at 8 microns by choosing an appropriate thickness: 1.1 mm for MgO and 0.54 mm for LiF. Also shown is the measured normal specular reflectance of a polished piece of MgO ceramic. (Abrasive used was 0.3 μm Al_2O_3.) Provided that the material is polished, the reststrahlen reflectance spectrum is quite similar to single crystal material. Work by Voltz shows that pressed MgO powders do not achieve a high reststrahlen reflectance.[6] Thus the high density of ceramic MgO (>90% of bulk density) is required to achieve high reflectance.

The spectral normal emittance of smooth, fully dense layers of MgO and LiF backed with a reflective layer is by Kirchhoff's law just unity less the reflectance shown in Figure 1. However, the spectral emittance of materials which scatter radiation, such as the MgO ceramic, cannot be deduced solely from specular reflectance measurements. The spectral emittance of painted surfaces was also desired for this work since these surfaces are almost blackbodies and are useful as comparison radiators. To obtain direct emittance measurements, a simple infrared bandpass filter radiometer was used. The radiometer viewed the samples while they were heated to 100°C. The spectral emissivity of the sample was estimated by interpolating between measured values for aluminum foil (~0.05) and published values for a certain paint.[7] This simple measurement technique is admittedly crude, but for our purposes it was quick and effective.

Several MgO ceramic samples and two painted surfaces were evaluated; the results are given in Table 1. For the MgO samples,the 8-14 micron emissivity is large and relatively insensitive to surface finish. In the 17-22 micron range, the surface emissivity is improved (reduced) as the surface is polished. According to the calculations, the only significant effect of moderate thickness variations should be emissivity changes in the range 7 to 9 microns because it is only in this range that the absorption length is of the order of 1 mm. This idea is confirmed by the second and third lines of Table 1, which show a thickness reduction from 1.1 mm to 0.3 mm reduces the 8.3 to 9.1 micron emissivity from 0.79 to 0.68. This result demonstrates that MgO thicknesses of the order of 1 mm are required to produce a high "window" emissivity. No filter was available with a passband in the 5 to 8 micron wavelength range, so it was not possible to directly verify the low emittance in this range. However, since the absorption index declines with decreasing wavelength in this range, the 5 to 8 micron emittance should be small (The absorption index at 7 microns is[5] only 1.1 $cm.^{-1}$).

Spectral emissivity measurements for two samples of white paint are also given in Table 1. "White paint I" has been used by our group in prior radiative cooling experiments.[9] "White paint II" is a thick coat of TiO_2 paint with high pigment concentration. It has been used here as a standard of comparison because of its slightly higher emissivity. Both white paints are clearly non-spectral emitters.

To test the performance of polished MgO as a selective radiator to the night sky, a panel with an area of approximately 0.5 m^2 was fabricated. Magnesium oxide ceramic plates 1.1 mm thick, polished on the topside, were placed on aluminum foil to form the radiator. The radiator was well insulated on the underside. It was covered to eliminate convective heat transfer with a 50 micron polyethylene film mounted 2.5 cm above the radiator. The performance of this panel was compared with an identical panel having white paint II as a radiator. Both panels could be heated electrically. The results, Figure 2, show that the MgO panel can achieve minimum temperatures about 3^{o}C cooler than the reference painted panel. This performance is superior to any other material we have tested. In particular it out performs aluminized polyvinyl fluoride films, which we have previously compared to the slightly less emissive white paint I.[9]

While the MgO ceramic is the best selective radiator tested, it does not produce temperature depressions as large as those theoretically possible. In the absence of non-radiative heat gains, an ideal selective radiator can produce temperature depressions below air temperature about 2.4 times as large as a blackbody radiator.[10] There are several reasons for this non-ideal behavior.

First, the optical properties of the radiator are not perfectly matched to the atmospheric spectrum; beyond 25 microns the thermal emittance is not as low as would be desirable.

Second, under the dry atmospheric conditions for our test (dewpoint temperatures about $0^{o}C$), a secondary atmospheric window forms in the 17-22 μm range. Figure 3 shows our measured values of the zenith sky emissivity in the 17-22 μm band as a function of dewpoint temperature. (Air temperature near ground level is used to convert measured radiances to apparent sky emissivities: The measured spectral radiance was normalized by the radiance which would have been measured if the atmosphere were a blackbody at air temperature.) The lower sky emissivity in the 17-22 μm band at low dewpoint temperatures is an advantage for a blackbody radiator, but no advantage for the MgO radiator, which has a low emissivity in this region. This appears to be the primary reason that when the panels are heated with 85 Wm^{-2}, the painted panel can remain cooler as shown in Fig. 2. (The 8-14 μm emissivities of the polished MgO plates and white paint II are almost identical.) When the atmosphere is more humid, the overall radiative cooling effect will be somewhat diminished but the relative performance of the MgO panel should improve further.

Finally, we consider the effect of the non-radiative heat gains on the lowest temperatures achievable. For the selective MgO radiator, these heat gains are more significant because the operating temperatures are lower. Also, the radiative heat gains outside the atmospheric window are small for selective radiators, so non-radiative heat gains have a larger relative impact on selective radiators than on non-selective radiators. In a test to examine the importance of the conductive heat gains of the radiator, a panel was fabricated with a 5 cm air gap, twice the usual 2.5 cm value. Atmospheric conditions were similar to the test already reported. In this panel the MgO radiator cooled to 5°C below the reference panel. This substantial increase from the 3°C value reported above indicates the importance of minimizing the non-radiative heat gains.

In summary, it has been shown that magnesium oxide in a ceramic form can be fabricated into a useful selective radiator which cools well below ambient temperatures. It is capable of reaching lower temperatures than selective radiators based on aluminized polyvinyl fluoride. The high solar reflectivity of MgO may facilitate the development of systems which produce radiative cooling during daylight hours. Finally, the similarity of the infrared optical constants of LiF to those of MgO suggests that it can be used in a similar fashion. Thus radiators may be fabricated with LiF and mixtures of LiF and MgO.

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Fig. 1 Computed normal specular reflectance for layers of MgO (1.1mm) and LiF (0.54mm) backed with a reflecting layer. The normal spectral emissivity is unity less the reflectance. Also shown is the measured normal specular reflectance of a polished MgO ceramic sample (1.1mm).

Fig. 2 Thermal test results.

Fig. 3 Monthly average zenith sky emissivities in the 17-22 μm band, for clear skies.[10] This data is consistent with the known properties of water vapor absorption. Calculated values shown are based on the LOWTRAN model[11] which does not currently include continuum H_2O emission in this spectral band. The continuum absorption is about the correct magnitude to explain the discrepancy.

Table 1 title: Measured normal spectral emissivities.

Sample			Wavelength band (μm) 8-14	8.3-9.1	14.0-15.8	17-22
MgO	ceramic	polished, 1.1mm	0.91	0.81	0.55	0.21
MgO	ceramic	buffed, 1.1mm	0.89	0.79	0.54	0.26
MgO	ceramic	buffed, 0.3mm	0.85	0.68	0.54	0.26
MgO	ceramic	unpolished 2mm	0.92	0.86	0.66	0.55
White paint I			0.88	0.83	0.91	0.87
White paint II			0.93	0.86	0.93	0.92

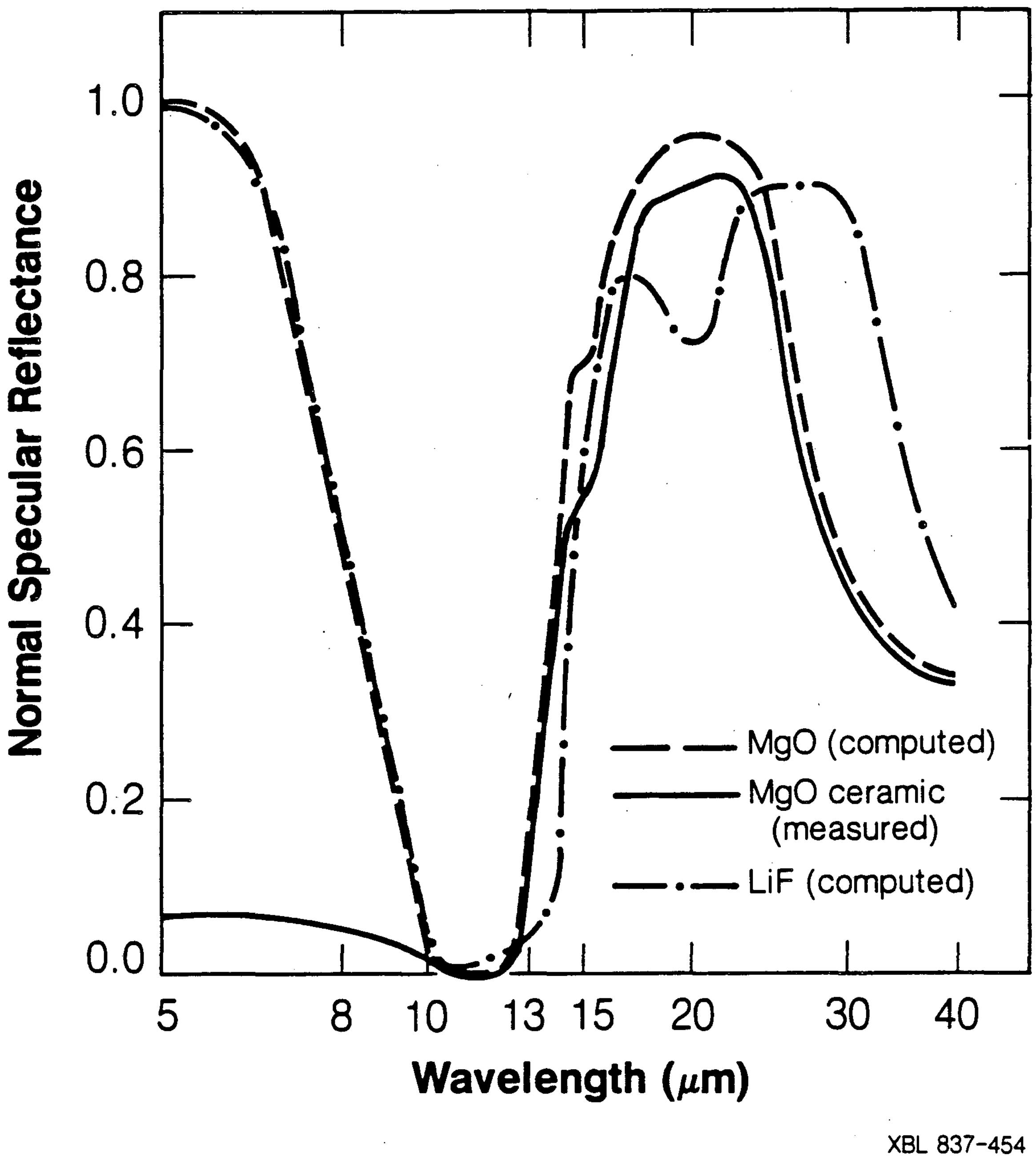
Normal Specular Reflectance
1.0
0.8
0.6
0.4
0.2
0.0
5
8
10
13
15
20
30
40
Wavelength (μm)
MgO (computed)
MgO ceramic (measured)
LiF (computed)
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Fig. 1

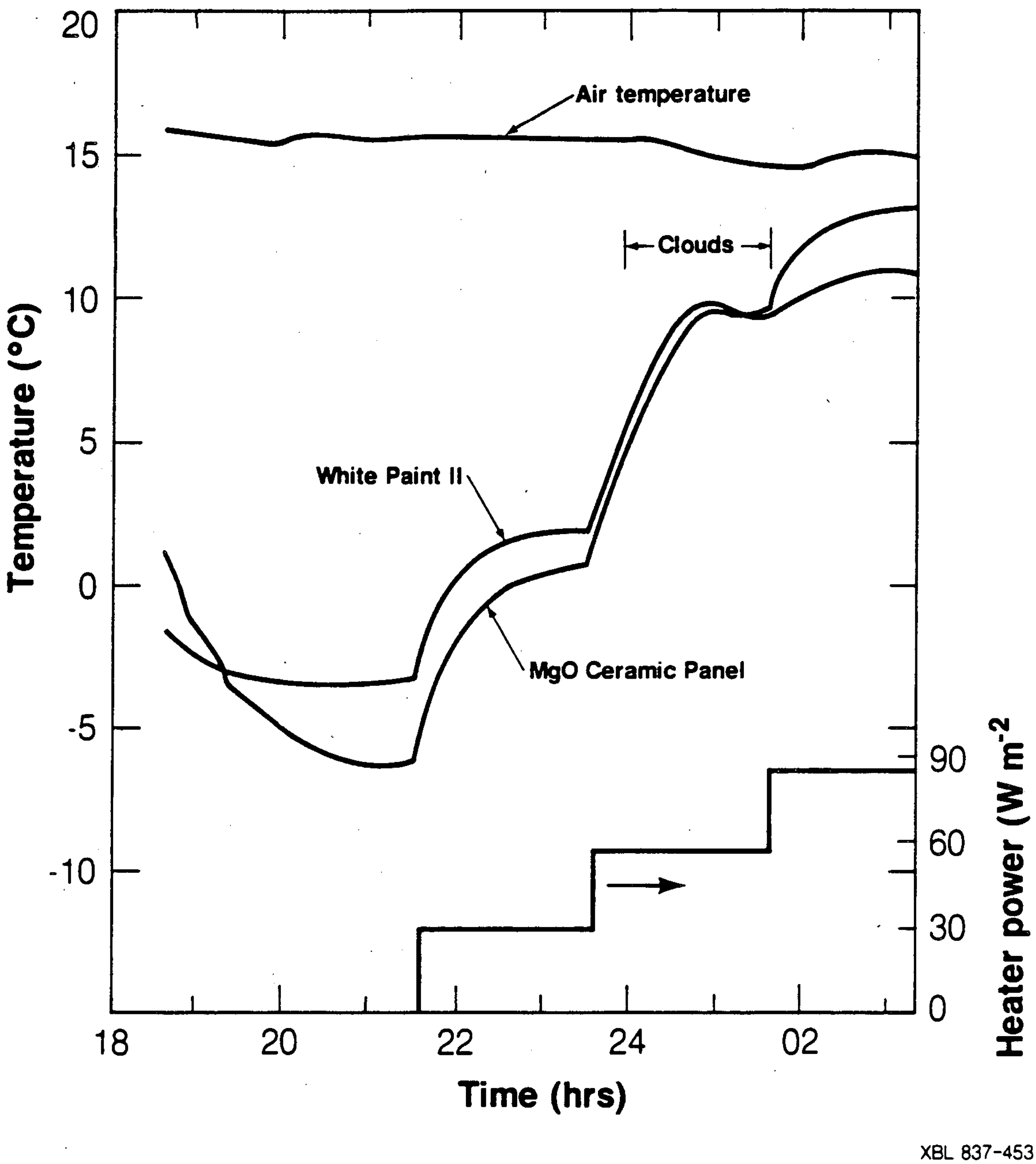
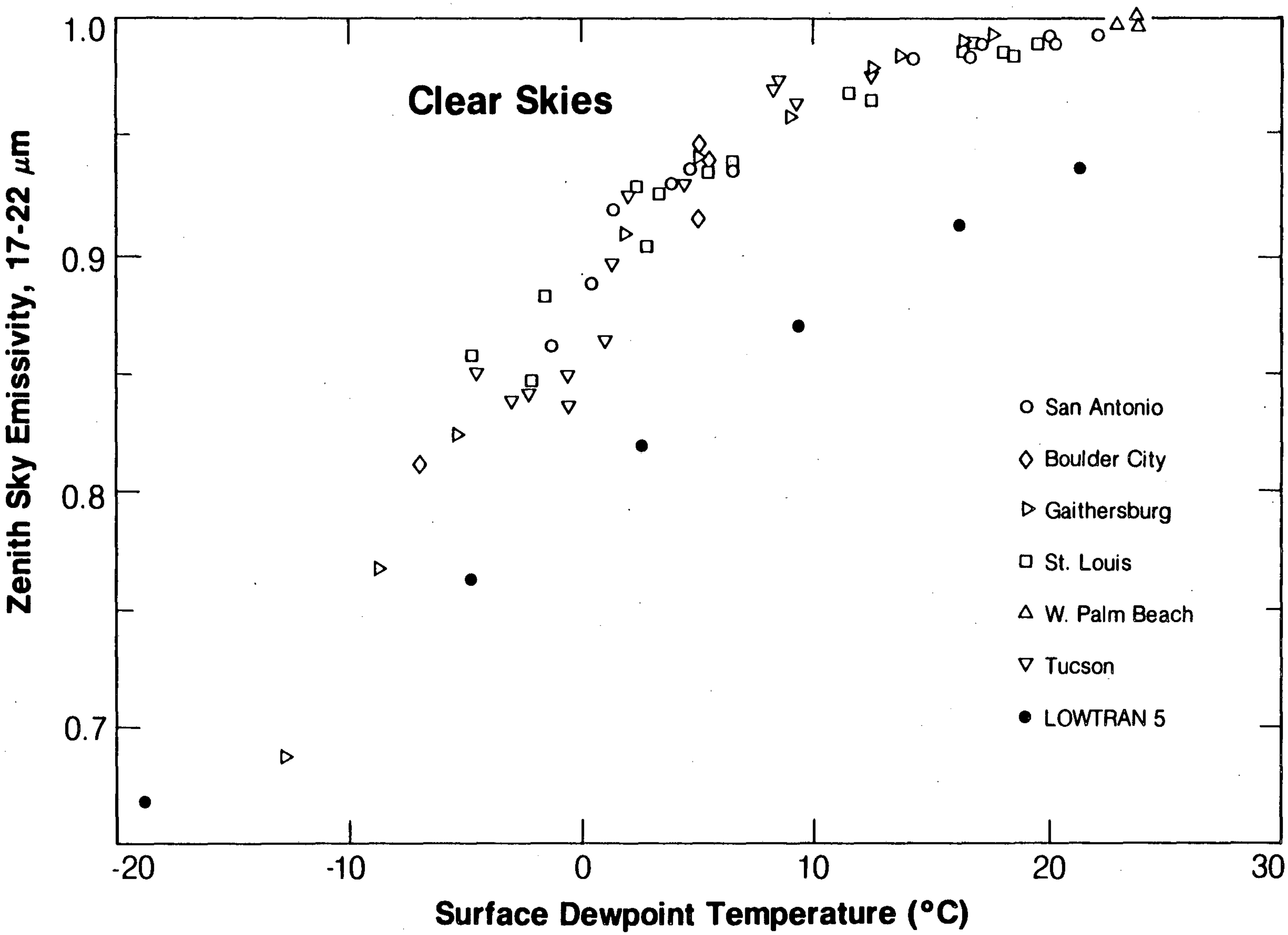

Air temperature
Clouds
White Paint II
MgO Ceramic Panel
20
15
10
5
0
-5
-10
Temperature (°C)
90
60
30
0
Heater power (W m-2)
18
20
22
24
02
Time (hrs)
XBL 837-453

Fig. 2

Fig. 3



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