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High Damage Threshold $Al_2O_3 \cdot H_2O$-SiO_2 HR Coatings Prepared by the Sol-Gel Process

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Our study of highly reflective (HR) dielectric coatings prepared by the sol-gel process has indicated that hydrated alumina and silica are the materials of choice for the high and low index components respectively.

We can now prepare 32-36 quarter-wave-layer samples from colloidal suspensions of the relevant oxides by spin coating at room temperature. These have reflectivities of about 99% and damage thresholds in the range 30-40 J/cm^2 at 1064 nm with 16 ns pulses at 30 Hz. This threefold improvement in threshold over that reported earlier for this system was attributed to an improvement in the preparative technique particularly in regard to cleanliness. Coatings have been prepared recently on substrates up to 8" in diameter and scale-up development continues.

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Introduction

The work described in this paper is part of a continuing study on the preparation of high damage threshold coatings suitable for use on the optical components in high power laser systems. Earlier investigations with the same objective on porous silica anti-reflective (AR) coatings [1], porous fluoride AR coatings [2], TiO_2 and TiO_2-SiO_2 high reflectivity (HR) coatings [3] and $Al_2O_3.H_2O$ and $Al_2O_3.H_2O$-SiO_2 HR coatings [4] have been previously reported.

Our investigation throughout has been based on the use of colloidal suspensions as coating media. These have the following advantages over other systems:

1. Liquid system which is easily applied at room temperature by conventional means.
2. No cure or heat required. Stress-free.
3. Rapid application
4. High purity product
5. Especially good for large substrates.
6. Low capital equipment cost.
7. Coating easily removed and replaced with no damage to substrate.

In our work with HR coatings we have always used SiO_2 as the low index component. Colloidal suspensions of SiO_2 are readily prepared and can be used to give high damage threshold, single layer SiO_2 coatings [1]. We have used these as AR coatings on substrates up to one meter in diameter. TiO_2, ZrO_2, HfO_2, Ta_2O_5, and AlO.OH were selected for investigation as the high index component. All but AlOOH were eventually rejected mainly because single coatings from each material had only low to moderate damage thresholds. The preparation of suitable colloidal suspensions of ZrO_2 and HfO_2 were also quite involved. These would have been difficult to prepare in quantities suitable for application to large substrates.

Suspensions of AlO.OH were found to be readily prepared and to give high damage threshold single layer coatings. The AlOOH-SiO_2 system was therefore chosen for further investigation. The initial work was carried out on 5 cm diameter substrates but our ultimate objective was to scale-up to much larger sizes.

Experimental Procedure

A. Colloidal Suspensions

Suspensions of AlOOH and SiO_2 were prepared by the hydrolysis of sec-butoxide aluminum and tetraethylsilicate respectively by methods previously described [1,4]. Both alkoxides were distilled prior to use to ensure a high purity product.

The SiO_2 suspension was prepared at 3% concentration in ethanol and then diluted to 2% with ethanol prior to use. The AlOOH suspension was prepared at 1% concentration in water. It was then adjusted to pH 5 with an ion exchange resin, evaporated under vacuum to 16% and finally diluted to 4% with methanol prior to use.

B. Coating Procedure

All coating suspensions were filtered through a 0.2 μm membrane filter. 5 cm diameter and 1 cm thick fused silica or BK-7 substrates were then coated using a spin coater in a 0.2 mm filtered, forced air horizontal flow clean hood. The first coat applied was always SiO_2 and this was followed with alternating AlOOH and SiO_2 coatings allowing about 10-15 minutes drying time between coats. The final coat was AlOOH. All coatings were prepared at an optical thickness of 266 nm corresponding to quarterwave at 1.06 μm.

The preparation and processing of both suspensions is illustrated in figures 1, 2 and 3.

C. Damage Threshold Measurements

Damage threshold measurements were carried out at 1064 nm using multishots at a pulse length of 16 ns at 30 Hz. The beam spot size was approximately 1 sq. mm. Each site on the sample was irradiated for 60 seconds (1800 shots) and then inspected for damage.

A new site was then selected and irradiation repeated. From a sequence of irradiations, the damage threshold was defined as the average of the highest fluence which caused no damage and the lowest fluence that did.

Discussion of Results

The refractive indices of the SiO_2 and AlO.OH prepared from colloidal suspensions have been shown to be 1.22 and 1.44 respectively [4]. A large number of alternating layers will therefore be required for high reflectance in a HR mirror system because of the low index difference. Calculations indicated that about 30-36 layers would be required to achieve reflectance greater than 99% on BK-7.

Table I shows the actual reflectances obtained for a variety of samples containing from 26 to 40 layers. It can be seen that at least 34 layers are required for 99% reflection. Figure 4 shows the transmission curve of a fused silica substrate coated with 36 layers. The reflectance of this sample is greater than 99% at 1064 nm. The narrow band third harmonic reflectance of about 95%, which is characteristic for this type of quarterwave stack is also present.

Laser damage threshold measurements at 1064 nm (16 ns pulse, 30 Hz) are also listed in Table I. These range from 21-50 J/cm^2 with an average of 37 J/cm^2 with no apparent dependence on number of layers. This perhaps is not unexpected as the first few layers in all samples are exposed to, and reflect most, of the fluence. These damage thresholds are much higher than we have reported previously [4]. While the reason for this is not known for sure we suspect that improvement in technique, especially in regard to cleanliness, has made a major contribution.

Our efforts are now directed towards scaling-up the coating process. We are currently coating 8" diameter fused silica substrates and have encountered a few minor problems. These include an increase in the number of artifacts and "comets" in the coatings and, in some cases, radial lines have appeared. The artifacts mean that we must improve our cleanliness to an even greater extent and the radial lines are probably due to non-uniform drying and suspension instability during coating. These factors are all under investigation.

Conclusions

We have selected AlO.OH - SiO_2 over several other candidates for continuing evaluation as an HR coating system.

Our research on small 2" diameter samples has indicated that this system has high laser damage threshold and adequate optical performance but requires a large number of layers (32-36).

A coating system for 8" diameter substrates is under development.

References:

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[2] Thomas, I. M. "Porous Fluoride Antireflective Coatings," Appl. Opt. 27, 3356 (1988).

[3] I. M. Thomas, "Single Layer TiO_2 and Multilayer TiO_2-SiO_2 Optical Coatings Prepared from Colloidal Suspensions," Appl. Opt. 26, 4688 (1987).

[4] Thomas, I. M. "Single Layer $Al_2O_3.H_2O$ and Multilayer $Al_2O_3.H_2O$-SiO_2 Optical Coatings Prepared from Colloidal Suspensions," Appl. Opt. 28, 4013 (1989).

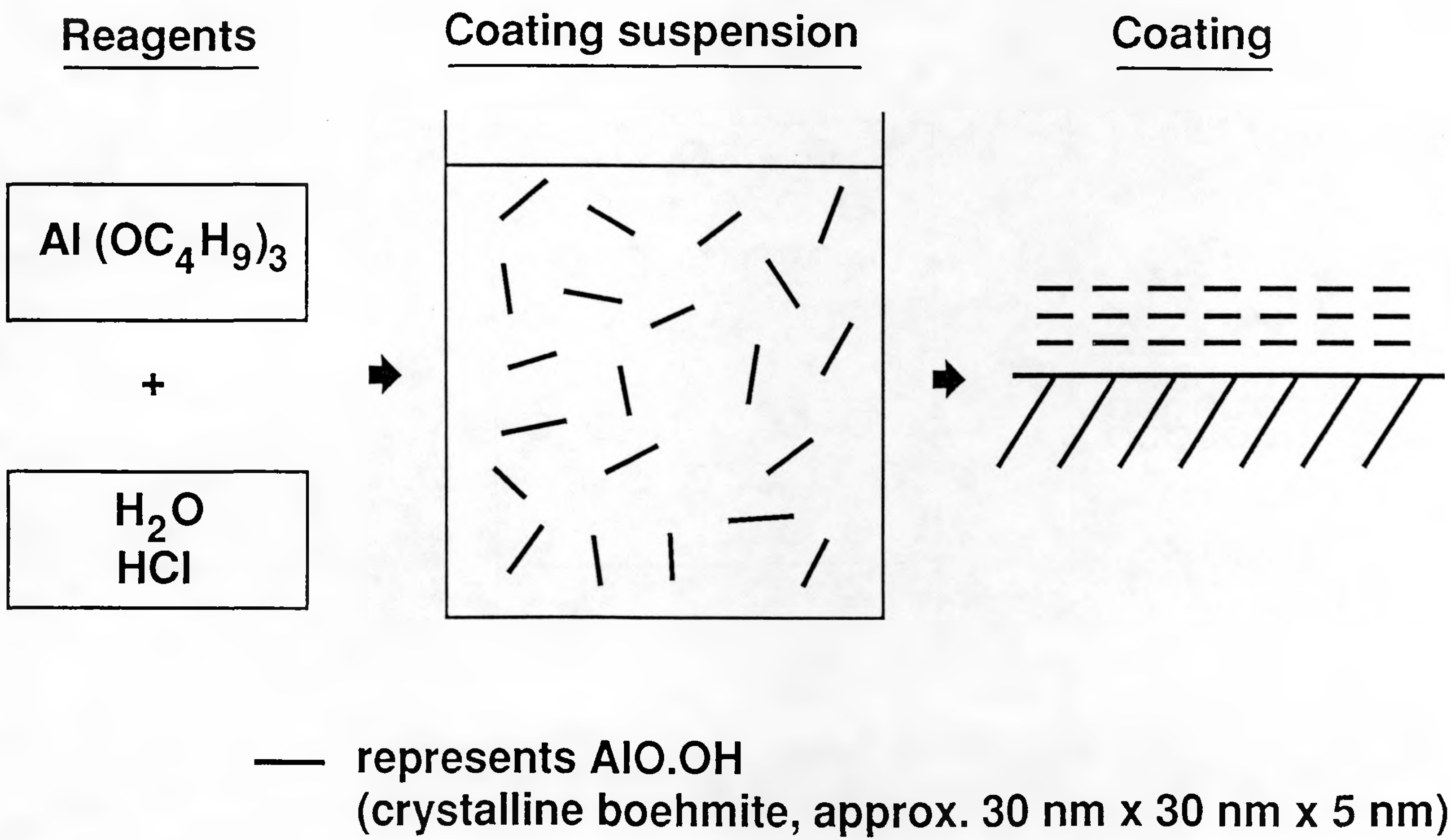


Fig. 1. Preparation and application of colloidal AlO.OH.

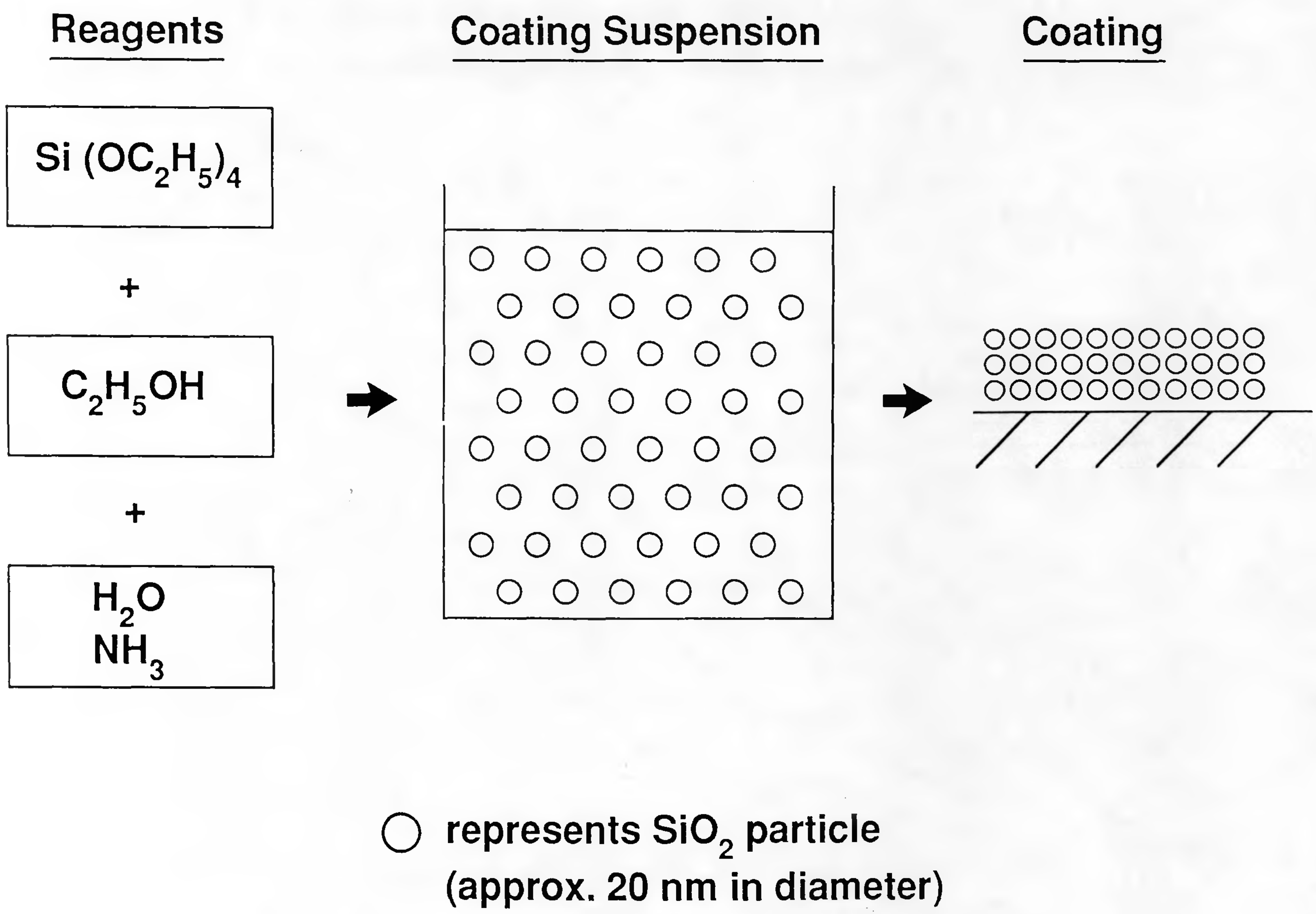


Fig 2. Preparation and application of colloidal SiO_2

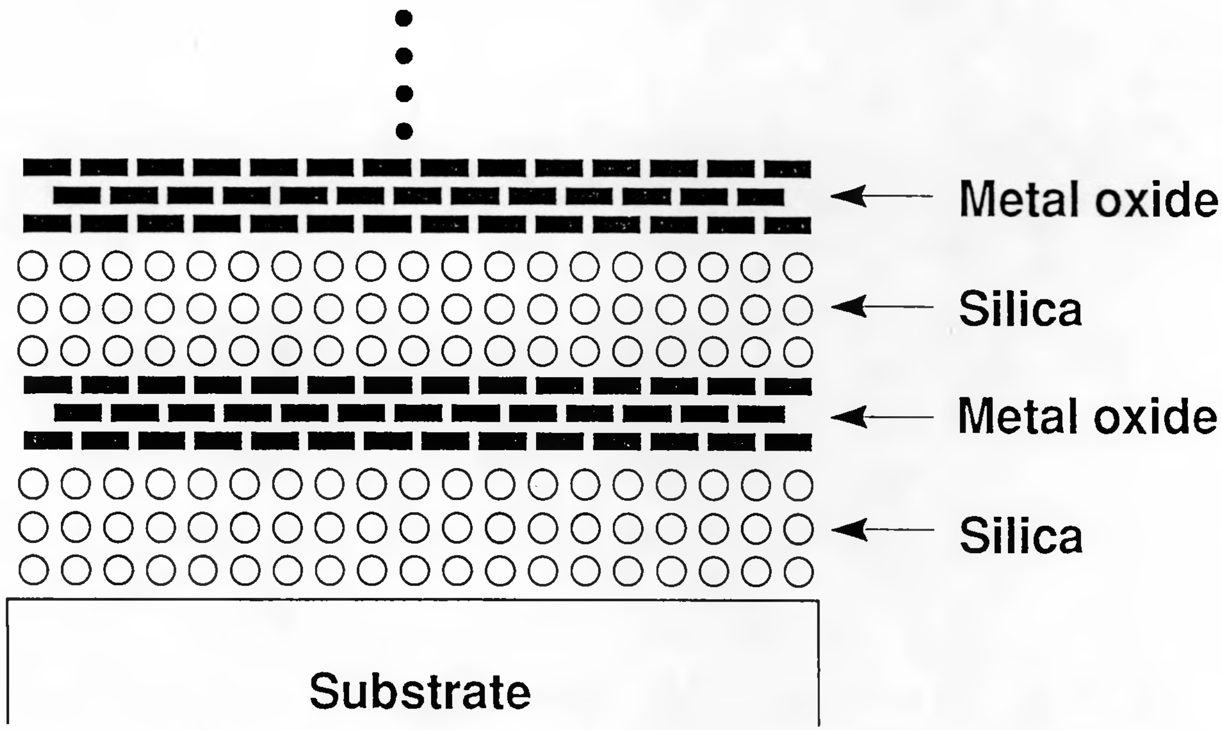
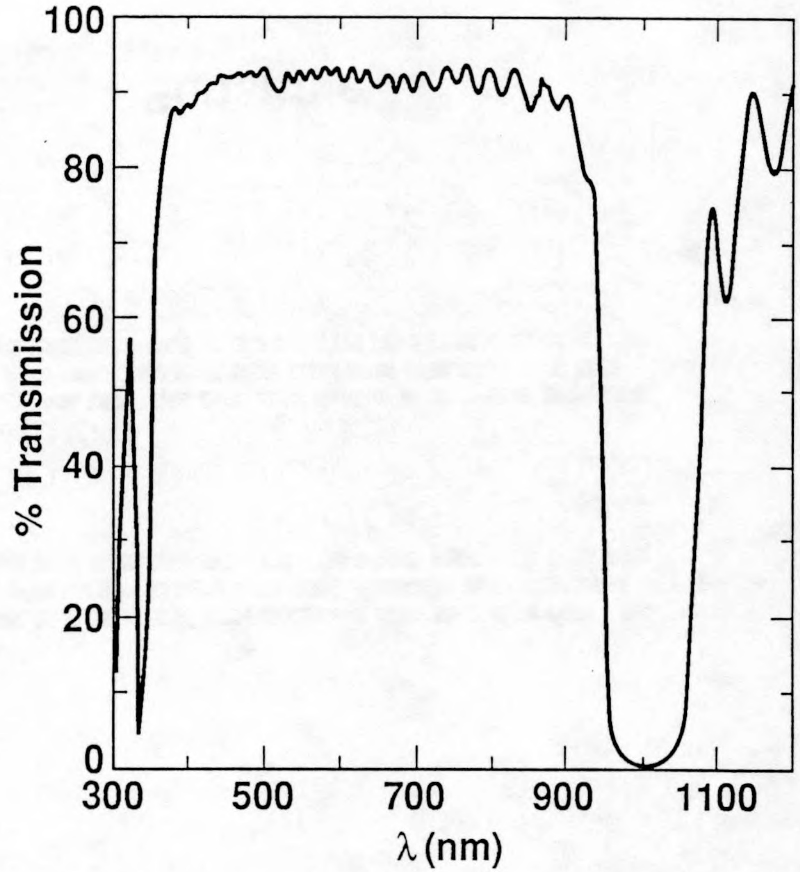


Fig 3. Multilayer HR coating



$[SiO_2\text{-}Al_2O_3]^{18}$

Fig 4. Transmission spectrum of 36 layer HR coating

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Layers	Reflection at 1.06 μm	Damage 1064 nm/16 ns
26	95.0 %	34 J/cm^2
30	98.0	45
30	98.5	28
32	98.0	38
32	98.5	50
34	99.0	21
36	99.0	37
40	99.5	40

Table 1. Reflectance and damage thresholds of several HR coatings.