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THE DEVELOPMENT AND APPLICATION OF POLYURETHANE COATINGS FOR HIGH REFLECTIVITY,

(10) A. S. DALTON

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AERONAUTICAL SYSTEMS DIVISION
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UNITED STATES AIR FORCE
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

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200 - May 1962 - 32-1432 & 1433

FOREWORD

This report was prepared by the ~~Materials Engineering Branch of~~ the ~~Applications Laboratory~~. The work was initiated under Project 7312, "Finishes and Materials Preservation," Task No. 73121, "Organic Protective Coatings," and includes data reported in WADC TR 55-258 dated April 1956 (Contract AF 33(616)-2317), WADC TR 56-489 dated September 1956 (Contract AF 33(616)-3095), and WADC TR 56-622 dated April 1957 (Contract AF 33(616)-2890).

Research started in April 1954 and completed October 1961.

This report presents a paper, the essential data of which were presented to the 26th meeting of the Surface Preservation Section, Production Techniques Division, American Ordnance Association at Santa Monica, California in October 1959. The meeting had controlled attendance although the paper was not presented as a classified document. This material, as amended, was released for publication by the Directorate of Security Review, Department of Defense on 2 May 1961.

Acknowledgement is made to Mr. R. L. Stout, who monitored the first two contracts above, and helped in consolidating and organizing the data reported herein.

ABSTRACT

The objective of this paper is to present the importance of thermal radiation control in preserving the structural integrity of a weapon system and to outline the history of the investigation and definition of the problem, the research and development involved in solving it and the application engineering in adapting the solution to practical use. Polyurethane coatings have many desirable properties, show outstanding performance, and for this problem are better than any other coating investigated to date.

PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:

W. P. Conrardy
WALTER P. CONRARDY
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Applications Laboratory
Directorate of Materials & Processes

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INTRODUCTION

The purpose of any coating is one or a combination of three; Protection, Aesthetics, Special Effects. From a technical point of view, the Air Force is interested in coatings primarily for the first and third purposes. However, since appearance, or the aesthetic value, is of great importance psychologically and politically, the Air Force devotes much attention to this aspect also.

The individual properties of coatings which are of Air Force interest might be classified under the following: Optical, Physical, Chemical, Electrical, and Nuclear. Optical properties might be considered as another group under the physical properties, as electrical and nuclear might be considered under both physical and chemical properties. However, each of these three groups are of sufficient breadth and importance that they should be classified independently. Of particular interest in this discussion are the physical, optical and chemical properties.

To cover adequately all of the coating problems to which polyurethane coatings might specifically apply would be rather impractical, if not completely impossible, in the relatively short time allotted. Therefore, outside of the brief introduction above to "set the stage," this paper is concerned with an investigation of certain properties of polyurethanes with an "eye" toward meeting a specific problem within the Air Force, the work involved in developing a satisfactory specification to cover the resultant material, and some of the many possible applications of this material, both present and future.

PROBLEM DEFINITION

As with any applied research program, this program had its special impetus. With the advent of atomic and thermo-nuclear weapons an entirely new, perplexing question revealed itself. This was the problem of what could be done about the structural damage to aircraft, caused by the intense thermal radiation. As are many problems, this one was discovered by accident and found us unprepared, especially for its magnitude. The terrific heat produced by these weapons severely damaged a number of aircraft before the extent of the trouble was determined.

Of course the greater the distance from the source, the less severe the damage. This distance, however, is limited by the speed and altitude of the aircraft, the configuration of the approach and escape, and the type of drop or effect desired. An additional factor that had to be considered was the size or yield of the weapon. This factor varies greatly and no established set of parameters could be made.

At the minimum distance, varying with the yield of the weapon, the spectral energy distribution from these blasts closely approximates that of the Sun. Since the vast majority of solar energy lies within the visible (0.4 to 0.7 microns) and the near infrared (to 2.0 microns) portions of the spectrum, a material with high apparent daylight reflectance will reflect most of the thermal radiation and thus prevent serious structural damage to the aircraft. The most expedient solution to the problem at hand, then, seemed to be to coat

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Manuscript released by the author 26 October 1961 for publication as an ASD Technical Documentary Report.

the vulnerable areas of the aircraft with white paint. Standard specification alkyd enamel was utilized since it was readily available and provided a rapid, if temporary and not optimum solution.

This material, being pressed into service where it was not specifically intended, failed in many cases. This was not necessarily from thermal degradation, but from softening, cracking and peeling because of conditions of atmospheric heat and moisture plus a certain lack of personnel education and care.

While exceptionally white and reflective initially, if manufactured and applied properly, standard specification alkyd enamels chalked badly on exposure and picked up soil rapidly causing a large drop in reflectivity. It became imperative to search for and develop a material which would resist the extreme condition of thermal radiation as well as maintain good general paint properties.

RESEARCH AND DEVELOPMENT

Even though high reflectivity from an optical point of view is the primary property desired, the ability to withstand high temperatures for short periods of time without charring or even darkening appreciably, and thus losing the good reflectance, is also important. The point at which this darkening and charring occurs represents the critical energy of the material. Purity of materials is paramount. Even a trace of tinting material proved to be a serious contaminant from the standpoint of obtaining higher critical energy.

Many materials were investigated: Various coatings, pigments, resins, extenders, and special additives. Their effects upon the critical energy of many coating systems were studied. Table 1 shows the critical energies of five different types of coatings, and the effect of the primer on the entire system. All of these coatings were pigmented at the same volume (except the specification enamel) and were applied over a specially formulated primer (except the one polyurethane over standard zinc chromate primer) on .016 aluminum alloy panels. The polyurethane has approximately 30 percent higher critical energy than three other good coatings and 800 percent higher than standard alkyd enamel. Also it is evident that a specially and properly formulated primer will increase the critical energy approximately 30 percent over standard zinc chromate primer, showing that a special primer is necessary. Work is completed on this special primer. A specification, MIL-C-27316(USAF) has been released. It covers an epoxy-polyamide based material.

The prime benefit to be gained with any thermally reflective system is a lower metal temperature. As can be seen from the data in Table 2, the polyurethane coating provides appreciably less temperature rise than other coatings of merit, and much less than standard alkyd enamel. Again a specially formulated primer provides another bonus, in less temperature rise, to the entire coating system.

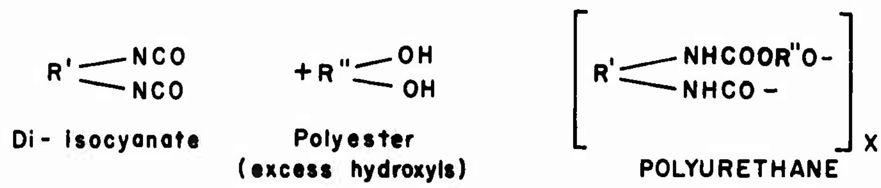
The critical energy and specific temperature rise data were compiled in the Material Laboratory, New York Naval Shipyard. These data were determined by means of the searchlight test on prepared specimens.

Other properties, though perhaps of somewhat less importance, were sought. Table 3 shows some of the more important ones in addition to those already discussed for four coatings.

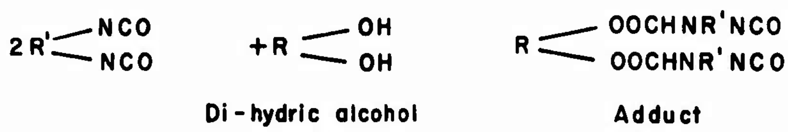
In the overall picture of generally important properties, the polyurethane is much better than the other three types of coatings which have shown considerable merit. It is most outstanding in its high critical energy, its low temperature rise, and its excellent resistance to synthetic lubricants and hydraulic fluids. In addition, it has excellent abrasion and erosion resistance, unusual adhesion to all types of substrates, good impact resistance, good water, solvent, and chemical resistance, and excellent electrical insulating properties. A special property of note is its low temperature flexibility (-65°C).

The vacuum volatility of these coatings at temperatures up to 300°F is very low, in the neighborhood of 2 to 3 percent weight loss. This is predicated on a properly cured coating.

As an introduction and refresher to those not entirely familiar with polyurethane coatings, the following general reaction is given:



The di-isocyanate in itself is quite volatile and toxic. In actual practice and production, an intermediate is first made similar to the following reaction:



This intermediate reaction gives a stable liquid reactant which is much easier to handle. These equations are merely symbolic of typical reactions. Many modifications are possible and many of these variations contribute essentially to the specific properties of the final coating.

A wide variety of other materials besides polyesters can be reacted with di-isocyanates to give useful finishes. Linear polyamides and polyester-amides may be used in place of polyesters. Other materials such as epoxy resins, copolymers of glycol mono acrylates and styrene, phenol-fatty acids, silane diols, and certain chlorinated resins may be reacted to give desired properties.

As an item of novel interest, coatings can be made by treating monoglycerides of various fatty acids with an aromatic di-isocyanate and cured by exposure to moisture or steam.

This presents another chemical property which must be considered in the development and ultimate application of polyurethane coatings. This is the sensitivity of these resins to moisture, with which they react quite rapidly. This reaction evolves carbon dioxide gas, which if not properly controlled will cause the film to bubble extensively over the entire coating surface. These coatings must of necessity be applied and cured under conditions of low humidity (<80% RH) and for many applications in a vacuum. This vacuum cure is especially beneficial for space-temperature applications, for humidity during curing seriously affects the later stability of these coatings in a vacuum.

There are other properties and characteristics of polyurethane coatings which can be troublesome. One which is especially troublesome is the sensitivity to ultra-violet radiation. Light of shorter wave lengths than 0.4 microns rapidly discolors the film. Certain additives had to be found to overcome this deficiency. The control of flow and grinding ease were also problems that received special attention. Difficulties with solvent release and "open time" had to be studied and solved.

The fact that a two-component system was necessary presented logistic and application problems and encountered personnel reticence toward its use. Proper package adjustment and field education has or will undoubtedly eliminate these difficulties and the accompanying human reluctance.

This item of field education deserves a bit of special emphasis at this point. The largest percentage of problems encountered with the use of protective coatings stems from either improper surface preparation or improper techniques. As more complicated coating formulations and application techniques are released to the field, the problems are expected to be compounded. Only by proper education and willingness to learn can most of these difficulties be overcome.

In Table 4 the composition of the two components is given. The two components are mixed one to one just prior to use. This ratio, while somewhat difficult to develop in the laboratory, alleviates many of the handling and mixing problems encountered with normal multi-component coatings in the field.

Titanium dioxide is the only prime pigment in this formulation. Most of the reflectance of the coating is provided by this ingredient. The greater the pigment volume concentration up to 60 (this is generally near the point of critical PVC) the greater the reflectance. This high PVC is not practical for external applications, however, since it causes the coatings to be very porous. Rapid soiling and staining from run down and carbon deposits follow with this situation. Not only does this discoloration occur rapidly, but it is very tenacious, being nearly impossible to remove. Lower reflectance and critical energy then result. The pigment volume concentration of this formulation is 28 as based on the polyester resin portion. This provides excellent reflectance, gloss, and cleaning ease. After catalyzation, TiO_2 constitutes 6.5 percent of the total formula by volume.

As the PVC is lowered, generally the settling of the pigment component increases. Stearated aluminum silicate, 1.5 percent by volume, was found to lessen this difficulty to the point of practicability.

A series of three polyester resins of varying plasticity were mixed to obtain the desired physical properties of flexibility, adhesion, hardness and others. This resin blend constitutes 20 percent of the total volume.

Ethyl acetate proved to be the best solvent for the polyester paste portion. This constitutes 7 percent of volume. Urethane grade solvent is essential. For, as discussed above, moisture which is generally present in common grades of these solvents will interfere seriously with the desired reactions. Toluene, 3 percent was used merely as a diluent in this formula.

To control flow and leveling properties, 0.2 percent by volume of cellulose acetate butyrate was added. This plus proper solvent and diluent adjustment has essentially eliminated the objectionable "orange-peel" effect of earlier formulations.

To curtail the yellowing from exposure to ultra-violet light, 0.2 percent of an absorber was added (in this case Uvinul D-50 from Antara Chemicals).

This polyester resin base paste is ground in a clean non-contaminated pebble mill. A grind of 7 (Hegman gauge) is necessary to insure good gloss and dispersion. The paste is thinned with a combination of acetates, cellosolve acetate 6.1 percent and ethyl acetate 5.5 percent. A portion of this thinner should be withheld for viscosity adjustment.

The catalyst component consists of 35 percent, by volume, toluene di-isocyanate thinned with 7.5 percent cellosolve acetate (urethane grade again) and diluted with 7.5 percent xylene. Xylene was used to slow down the evaporation and setting time to allow for easier handling and application. Material resulting from this development program and compounded as outlined is covered by Military Specification MIL-C-27227(USAF).

APPLICATION

The primary objective in this entire development program is the protection of our bomber fleet.

This coating, after service evaluations of over 36 months on a high performance aircraft, is still holding up very well. It has resisted erosion and a variety of environmental conditions. It has yellowed somewhat more than desirable, but this difficulty has been lessened with the inclusion of the ultra-violet absorber in the final formulation. The coating system has since been applied to other production and Mod-IRAN aircraft, most recently in May and August of 1961. Initial reports indicate excellent performance.

Problems with application techniques, personnel education, and frequent resistance to progress must still be overcome on some fronts. However, it would appear from service data collected thus far that polyurethane based coatings have the best chance of satisfactorily meeting the aesthetic and practical coating standards required by USAF and yet maintain high reflectivity, possess high critical energy and provide low metal temperature rise.

There are other applications for which this coating has or will prove advantageous. Coating the top of the fuselage of tanker, cargo, and personnel aircraft with reflective paint provides lower fuel temperatures, lower temperatures for sensitive equipment and materials, and more comfortable conditions for passengers. Since the energy curves for the Sun and atomic heat (at the distance specified) are nearly identical, the background and developments of this program are applicable to problems of solar reflectance.

The excellent abrasion, erosion, fuel and lubricant resistance of these coatings make them a natural for ground support equipment. White coatings similar to the one which has been developed for aircraft application will provide cooler temperatures in cargo and personnel carriers and fuel supply vehicles. The U. S. Army has developed a similar material for application to portable water tanks.

Polyurethane coatings are finding use in space and missile applications and their future appears even brighter. Primarily for visual mating with strategic aircraft, the Hound Dog missile is being finished with the described coating. Other missile system offices are considering its use also.

Based on the excellent performance and unusual properties, a coating formulated as described previously, was recommended for and applied to one of the Transit series satellites. With more sophisticated techniques, more pure materials, and progressive compounding, coatings based on polyurethane resins will find more applications on satellites, space stations and space ships in the future.

SUMMARY

This is one specific example of a problem, perhaps peculiar to the Air Force being investigated, defined, researched, solved and applied in service. Out of it has grown many other applications, some related, but others unrelated to this specific problem.

This discussion has shown some of the many benefits to be gained from the use of coatings for protection as well as appearance. Polyurethane coatings show outstanding performance and are generally, for the purposes outlined, better than any other coating investigated. They have certain deficiencies, many of which have been or must yet be overcome.

It is not the intent of this paper to convey the impression to anyone that polyurethanes are the panacea of the coatings industry; but consideration of the problems, the parameters, the properties, the performance, and the ground rules strongly indicates that they have the best chance of satisfactory service for many applications and especially for the control of thermal energy resulting from the employment of atomic or thermo-nuclear weapons.

TABLE 1

CRITICAL EXPOSURE

IRRADIANCE - 10 $cal/cm^2/sec$
(cal/cm^2)

COATINGS OVER SPECIAL PRIMER	BLISTERS	YELLOWS	CHARS
SILICONE ALKYD	NONE	50	96
EPOXY ESTER	NONE	62	87
POLYURETHANE	NONE	53	124
POLYURETHANE (zinc chromate primer)	NONE	—	93
ACRYLIC	27	66	94
ALKYD (MIL-E-7729)	15		

TABLE 2

TEMPERATURE RISE

.016 ALUMINUM COATINGS OVER SPECIAL PRIMER	IRRADIANCE - 10 $cal/cm^2/sec$ $°C/cal/cm^2$
SILICONE ALKYD	5.2
EPOXY ESTER	5.2
POLYURETHANE	4.8
POLYURETHANE (zinc chromate primer)	5.2
ACRYLIC	5.9
ALKYD (MIL-E-7729)	12.0

TABLE 3

GENERAL PROPERTIES

	ACRYLIC	EPOXY	SILICONE ALKYD	POLYURETHANE
INITIAL REFLECTANCE	92.2	91.8	91.6	91.0
REFLECTANCE 500 °F/30 Min.	63.3	23.2	41.6	56.6
CRITICAL EXPOSURE (Cal/cm^2)	94	87	96	124
TEMPERATURE RISE ($°C/cal/cm^2$)	5.9	5.2	5.2	4.8
SOIL RESISTANCE	EXCELLENT	FAIR	GOOD	EXCELLENT
CLEANABILITY (percent initial reflectance)	92 %	85%	89 %	93 %
DRYING TIME	1 Hr.	1 Hr.	2 Hr.	1 Hr.
SPECULAR GLOSS	86	90	82	94
FLEXIBILITY 1/8" MANDREL	SLIGHT CRACKING	SLIGHT CRACKING	VERY SLIGHT CRACKING	VERY SLIGHT CRACKING
ADHESION	EXCELLENT	EXCELLENT	EXCELLENT	EXCELLENT
REMOVABILITY	FAIR	DIFFICULT	EASY	FAIR
SYNTHETIC LUBRICANT RESISTANCE	POOR	FAIR	GOOD	EXCELLENT

TABLE 4

COATING COMPOSITION

POLYESTER RESIN BASE COMPONENT	VOLUME PERCENT
TiO_2	6.5
STEARATED ALUMINUM SILICATE	1.5
POLYESTER RESINS	20.0
ETHYL ACETATE (urethane grade)	7.0
TOLUENE	3.0
CELLULOSE ACETATE BUTYRATE	0.2
UVINAL D-50	0.2
THINNER PORTION	
CELLOSOLVE ACETATE	6.1
ETHYL ACETATE	5.5
DI-ISOCYANATE CATALYST COMPONENT	
CELLOSOLVE ACETATE (urethane grade)	7.5
DI-ISOCYANATE RESIN	35.0
XYLENE	7.5

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