

Advances in Alkyd Resin based Paints from Prehistoric Era to Recent Time of Nano Coatings

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Abstract— As per historic assumptions, a painting art that is approx 5000 years old, the ancient Egyptian civilization gave shape to an extremely symbolic and fascinating era of paintings and sculptures that mesmerize people to this day. In prehistoric era, paints might have been prepared with natural gums, natural resins, plant-extracts, egg-extracts, animal's fat etc as binder whereas grinded rock powder & minerals as pigment and water had been as a common solvent. Paint preparations have been improved gradually as per developments of civilization by applying innovative techniques and technology to get superior quality of paints and surface coatings. Systematic study has made known that contemporary paints are being formulated to encompass shape from amorphous to crystalline, from micron size to nano size, configuration, dispersion stability, film smoothness, better aesthetic looks, required enhanced quality and forcefulness to both exterior and interior paintings of industrial automobiles by applications and blend of appropriate ingredients i.e., resin, solvents, additives and pigment.

In progressive circumstances, paint technology presents an extensive and wide range of methods and technologies that design and make advances in the exterior as well as interior properties and characteristics of materials and components by the way of alteration, arrangement and coatings for the purpose of protection and decoration. Functions of the surface coatings technology have been extremely widespread i.e., from micro to nano technology and from extremely hard materials to soft polymeric coating materials. Recent novel applications of nano-materials in paint-formulations promise to carry out the requirement and fulfillment of automotive glossy as well as protective surfaces; calculated as 90-94%, enhanced adhesion; i.e. 100%, weathering resistance; (408-1416 hours passed in quick ultraviolet accelerated weathering resistance test), corrosion protection; (600-2400 hours passed in salt spray test) and get enhanced mechanical properties. Self-cleaning properties of painted surfaces have been attained for better protection by applying nano materials.

Index Terms— Surface protection, alkyd paints, nano coatings, adhesion, glossy finish.

I. INTRODUCTION

Even more remarkable, if we talk about paints of ancient-era, is the fact that most elements of Egyptians-art remained remarkably undamaged and stable over the period of 3000 years that saw no significant outside influence on the-then Nile valley civilization. From some of the most beautiful depictions of ancient wars, to the significantly symbolic representation of ancient religions, rituals and deities, the art of ancient Egyptian paintings have stood the test of time and

survived to this age; giving the recollection of a past era. Surface coatings technology has been commenced as surface science and engineering, corrosion engineering and paint technology in industrial areas [1,2,3,4].

In the modern and fast life-style, universal communications take place by automotive vehicles, trains, ships, and aero-space vehicles etc; that's why best possible protection of vehicles alongwith superior visual appearance are required mainly through their coated or painted surfaces. It is significant to point out that the bare structural surfaces are very much aggressive to corroding environment and also not generally for the purpose of good looking; therefore surface coatings are required to improve surface properties, protection of existing materials and products [4, 5]. Systematic selections of proper surface treatments are always based on acquiring a complete set of requirements on the surfaces of parts and components with respect to intended operating conditions. The substrate-surfaces are responsible for all mechanical, thermal, chemical, and electrochemical interactions with the environment. These lead to the most important functions that require fulfilling by performing surfaces such as erosion & corrosion resistant surfaces, glossy surfaces, decorative appearance, self-cleaning property, weathering resistance, wear and tear resistance, optical behavior, and corresponding interfacial activities for better adhesion and paint-bonding [5,6,7].

If progressive sequences of surface-preparation are considered, pretreatment or surface treatment line is established for surface cleaning and surface-modification by phosphating for the purpose of further steps of surface coatings on materials and components. To design for a suitable surface treatment process before coating is a tough task for a given combination of different corrosive material-surfaces. It is difficult to precisely and thoroughly understand the operating conditions of a part and a very large variety of workable materials and technological processes, are also to be well thought-out [4,8,9].

General estimates indicate that there are a large number of materials used in materials technology. Also, together with surface technology, more than 50,000 materials and 800-1000 different processes are practiced in India. Moderately, the process of selecting the suitable coating or surface treatment requires a systematic approach. The selection process needs to be implemented at an early stage of product development.

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It is necessary that developers already consider surface requirements during concept phases, directly after taking down customer and market demands. Based on the given operating conditions, three fundamental aspects should be clarified scientifically [8,9,10,11]:-

Aspect 1. Condition of outside surfaces:

- a) Functional characteristics of the outside surfaces.
- b) To establish the requirements of surfaces.

Aspect 2. Limitations and constraints for boundary conditions for the view of

- a) Technical point
- b) Trade and industry
- c) Design-to-cost perception
- d) Environmental issues
- e) Cost estimation of life-span of materials

Aspect 3. Selection of alternatives for best possible operations.

Progress in the methodology of contemporary surface treatment and surface coatings: in prehistoric era, rubbing or polishing of the surfaces, wiping and water rinse were followed as surface treatment before surface coatings. Following the above views i.e., Job of outside surfaces, limitations and constraints for boundary conditions, and selection of alternatives for best possible operation and standard practices for surface-treatment processes are being followed as per ASTM/BIS practices and as per requirements of modern automotive industries [12,13]:

- 1) Degreasing – 1
- 2) Degreasing – 2
- 3) Water rinsing - 1
- 4) Water rinsing - 2,
- 5) Derusting
- 6) Water rinsing - 1
- 7) Water rinsing - 2
- 8) Activation
- 9) Phosphating
- 10) Water rinse
- 11) Passivation (as per requirement)
- 12) Drying

Subsequent to completion of surface-treatment processes, primer-coat application (by dip coating/brush painting/spray coating/electrodeposition coating) is done before intermediate coat (i.e. middle coat). After completion of primer coating and intermediate coating, top coat is applied [9,10,12,13].

In the perspective of current progress in paint coatings for advanced shiny finish and glossy-looks, paint applications

may be differentiated between “thick” and “thin” film technology in reference with dry-film-thickness. Thick film technology stands for dry-film-thicknesses above 10 micro meter and thin film technology for the dry-film-thicknesses between 1 nano meter and 10 micron. However, this division is not fixed. The automobile industry is one of the most significant consumers of recent advances in the areas of surface coatings and nano technology as well. Coating industries are growing day by day in India and around the globe [8,,10,11,12].

In present era, coating not only provides the purpose of decoration but also the resources of protection of valuable metals, materials, structures and buildings from corrosion which almost accounts for 4% of world’s gross national product (GNP). All major paint and coating companies are investing huge amounts on their research and development (R&D) sectors to formulate specific paints, which should be compatible and suitable for today’s hostile environment. Although, many new formulations of paints and coatings have appeared to glow in recent years, but none is able to carry out all the requirements in a single paint-formulation [2, 5,9].

Industrial glossy multifunctional coatings provide protection against wind, hot, cold, rain and also against thermal impact from the sun and environmental pollutions. Comfort and safety functions such as sun-control, aging, anti-reflex and easy-to-clean surface properties are faced by functional coatings which are being deposited using advanced thin film technologies [5-9]. Coatings can either be soft or hard. Soft coatings are based on thin metal films embedded in dielectric layers for optical reasons. These soft and sensitive stacks have to be protected and thus, they are integrated in laminated glazing. Hard coatings consist of more robust ceramic materials or thicker metal films. They withstand mechanical wear and corrosive attacks to a certain extent and can be applied on the inside or outside of the glossy surfaces [10,11,14].

Very much significant examples of outside coatings are hydrophobic i.e. easy-to-clean coatings for enhanced optical performance while driving in rain. The use of light weight complex shaped polycarbonate glazing will be possible with appropriate UV protective anti-scratch coatings [6,19,20]. Future car generations will be equipped with transparent or head-up displays in order to magnify traffic signs and other critical events according to eye-tracking systems [21,22,23]. Smart coatings will be used to adjust the transmittance of the glazing to minimize the heat load and to protect the driver’s and passenger’s isolation [24].

Nano paints and coatings are such materials, produced by reducing the materials/particles’ size at the nano meter level or molecular level to form a viscous liquefied product. The appearance and usefulness of nanoparticles transport many

advantages and opportunities to paint and coating industries. Coating industry is the first among all to acquire the potential of nanotechnology. Addition of nanoparticles to paints and coatings can upgrade a lot of properties of coating system and can produce multipurpose coatings with a little cost difference. Such coatings, sometimes made of self-assembling mono layers are applicable in many ways; from scratch resistant coatings on glass to self-cleaning surfaces to moisture absorbing to anti-graffiti and corrosion resistance coatings. Further, unique composition, better strength and flexibility along with excellent gloss and transparency makes nano-coatings even more effective. Many of the nanoparticles like nano ZnO are non-toxic in nature and thus include an additional improvement to coating industries [2,14,24, 25].

II. MATERIALS AND METHODS

Nano particles have been procured by BYK Additives and Instruments Co. Ltd. And all other materials, formulations and testing facilities were made available by Berger Paints Limited, Kolkata. In this extensive progression of paint formulation work: five types of paints have been prepared: 1st micron sized TiO₂ pigment based alkyd-amino paint (butylated melamine formaldehyde resin), and nano pigment particles (Nano SiO₂: 0%, 0.5%, 1.0%, 1.5%, and 2.0% alongwith required micron TiO₂ pigment) modified alkyd-amino stoving top coats were formulated for the purpose of protection of automobile grade steel surfaces.

In this formulation, resin, solvent and additives were kept constant; pigments were manipulated for comparative study. All five types of paints were characterized as per known standard and then these were applied on different standard panels (15cm X 7.5cm, thickness-1mm) and cured at 130^oC for 30 minutes. All coated panels were cooled at room temperature then testing-processes were carried out as per standard practice.

III. RESULTS AND DISCUSSION

Ensuing surface modification by surface treatment and phosphating, surface coatings were applied.

Detail comparative evaluation was carried out. Satisfactory results have been found (given in table 1 and 2) as per following ASTM / BIS Quality test [12,13]:

- Gloss test (ASTM-D-523: measured at 60 degree angle of Glossometer; table 1):
90 % at micron sized particles surfaces,
91-94% at nano-pigment modified surfaces.
- Adhesion tests (ASTM- D 3359):
Passed 100% in cross cut adhesion test (table-1).

- Q.U.V. accelerated weathering test (ASTM- D 4587; table-2):
300 hours passed: micron sized particles based coated surface (on bare steel panels).
408 hours passed: micron sized particles-surface,
800-1416 hours passed: nano-pigment modified surfaces [12,13].
- Salt Spray test (ASTM-B 117: table-2.):
400 hours passed: micron sized particles-surface, on bare steel panels.
600 hours passed: micron sized particles-surfaces, with primer.
1200-2400 hours passed: nano-pigment modified paint-surfaces with primer and intermediate coats [12,13].
- Immersion test (ASTM- D 3912: in water):
Passed: the same as the salt spray test: 600-2400 hours.
- Aging test (ASTM- G 154): - It was performed at 80^oC for 48 hours in diesel/engine oils; there was no appreciable change on coated steel panels.
- DFT (ASTM- B 487) - DFT was kept as:
3-5 micron for phosphating
15-25 micron for CED primer
25-30 micron for surface
25-30 micron for top coat.

The applications of nano materials in paint formulations assure to fulfill this desire of improvement in surface protection. Figure 1 shows the representation diagram of conventional stoving paints and Fig. 2 the representation diagram of nano-coatings

Table 1. Test observations (gloss and adhesion test) of coated panels:

Paints: Pigments ratio (Total pigment 23% by wt. including nano)	Cross cut Adhesion (%)	Gloss at 60 ^o angle (%)
Micron pigment 23% by weight (Nano = 0%)	100/100	90
Included Nano Pigment 0.5 %	100/100	91
Nano 1.0 %	100/100	92
Nano 1.5 %	100/100	93
Nano 2.0 %	100/100	94

Table 2. Salt spray test and QUV test observations of coated panels:

Paints: Pigments ratio (Total pigment 23% by wt. including nano)	Salt spray test passed in hours	QUV Weathering test passed in hours
Micron pigment 23% by weight	600	408
Included Silica Nano Pigment 0.5 %	1200	800
Silica Nano pig. 1.0 %	1600	1000
Silica Nano pig. 1.5 %	2000	1200
Silica Nano pig. 2.0 %	2400	1416

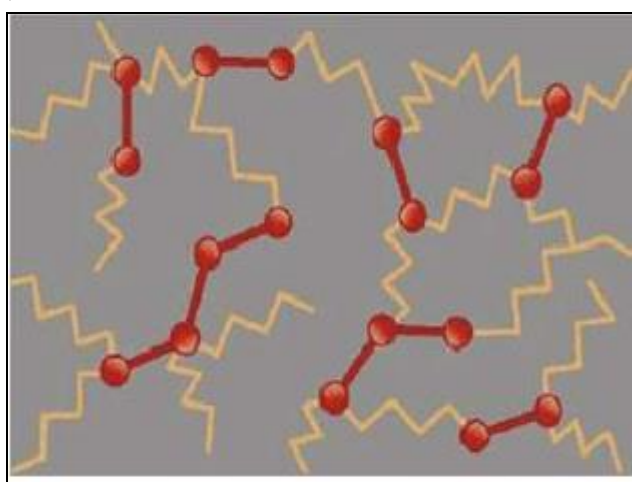


Figure 1. Automotive stoving paints consist of binder (resins such as alkyd, polyester, acrylic etc: shown in yellow colour) and cross-linking agents (such as amino resins: shown in red colour).

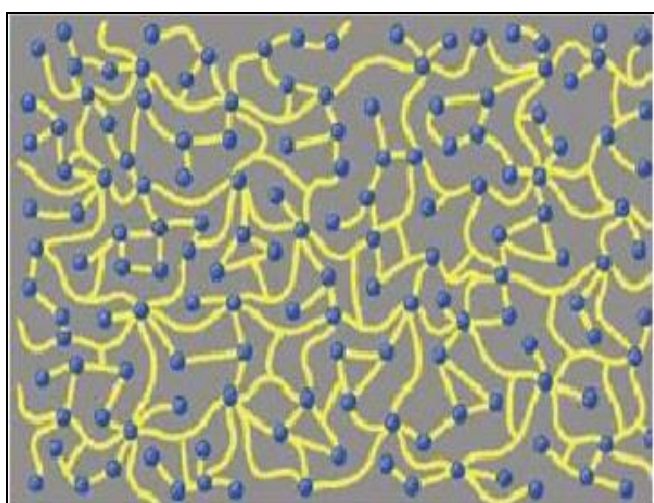


Figure 2. Nano coatings containing organic binder with high elasticity (in yellow colour) and inorganic nano particles with high surface energy (in blue colour).

Characteristic applications of functional coatings for the purpose of corrosion protection of metallic surfaces have been shown in figure 3. Surface technology focuses on reacting adequately to the specific kinds of load and stress. For this, the materials properties of surfaces are systematically modified particularly by means of volume loads and surface loads. A common summary of the most important processes is offered, along with their individual assets and drawbacks. Surface-modification processes for applications of paint-coatings involve covering the surface of a work piece with a well-bonded layer of shapeless material. Bond strength to the substrate material primarily determines the quality of a coating. This macroscopic property is controlled by proper combination of materials, types of interfacial zone, microstructure and process conditions, substrate-types and surface-treatment of substrate-surfaces. Corrosion resistance of coated samples was also examined by a neutral salt spray test according to ASTM B-117 specifications.

Salt spray test provides information about the nature of paint-failures like blistering, rusting, cracking, chalking and de-lamination of coating films. Corrosion protection properties of the sol-gel coated mild steel substrates were evaluated by exposing the substrates to a salt fog chamber having 3.5 % aqueous NaCl solution at 27°C atmospheric temperature [12,13].

The salt spray test image showed that after 1200 hours, the 1.5 weight% and 2.0 wt% nano-materials modified coating panels do not corrode while the other formulation samples corrode severely. This may be due to minimization in film-permeability, high surface energy, and higher contact angle result lower corrosion rate by 1.5 & 2.0 wt% nano coating samples compared to others.

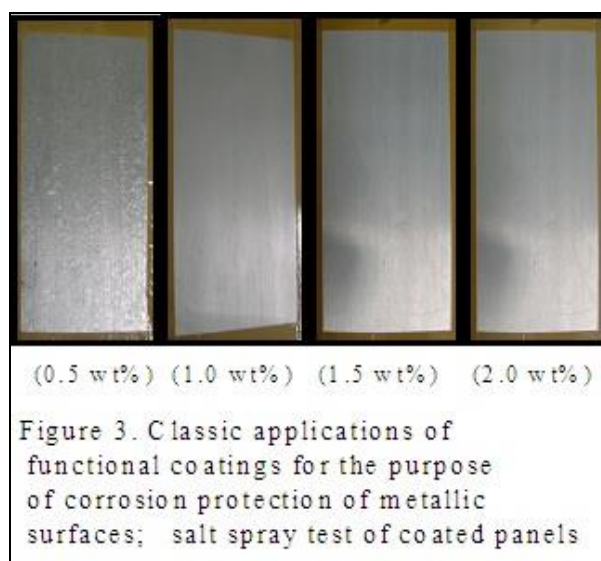


Figure 3. Classic applications of functional coatings for the purpose of corrosion protection of metallic surfaces; salt spray test of coated panels

A strong molecular bond in the contact zone has been most favourable; provided internal stress within the coatings, are

not too high and therefore no long-term degradation occurs within the coatings/substrate composite. Coating process and materials combinations determine whether mechanical, chemical, or electrostatic bonds prevail, or whether diffusion occurs. Thus, preparation of the work piece is a crucial factor in obtaining good coating adhesion. Apart from removing contamination, pre-treatments activate the substrate surface and therefore substantially influence the bond between coating and substrate. Classic mechanisms of surface activation are:

- To abolish oxide layers from the surfaces
- To make etches onto the substrate surface
- To enhance surface energy
- To make active surface molecules for further treatment i.e. phosphating

Quick ultraviolet accelerated weathering resistance test

(QUV test): Weathering degradation caused by UV rays is a common cause of paint failures of most of the coating systems. It causes the oxidation and decomposition of polymer films along with inorganic or organic pigments. Organic UV stabilizers also undergo deterioration after certain periods. Using nanoparticles like titanium dioxide or zinc oxide improve UV resistance property by not only absorbing but also reflecting those harmful rays. Also, they are not easily destroyed by UV rays and hence can increase the life span and weather resistance of paints [11,12,25].

Scratch resistance: Scratch resistance of coating can be improved by using micron sized inorganic fillers, but they cause matt or semi-matt appearance to coating by scattering visible light.

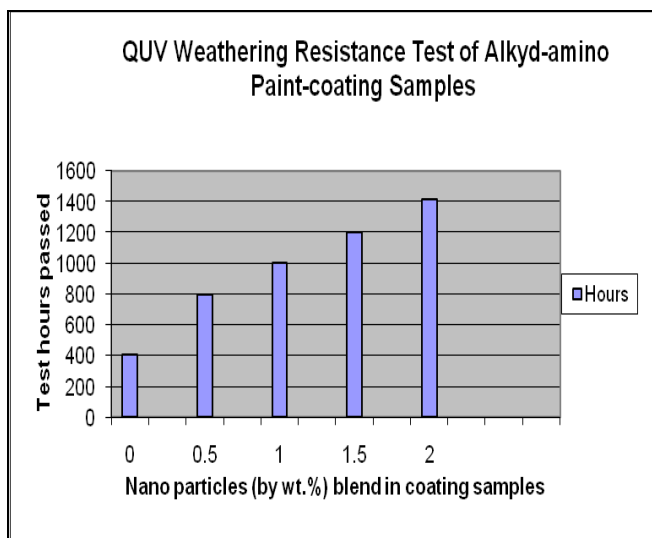


Figure 4. QUV weathering-resistance test of painted samples; weight % ratio (0, 0.5, 1.0, 1.5, 2.0) of SiO₂ nano pigment particles.

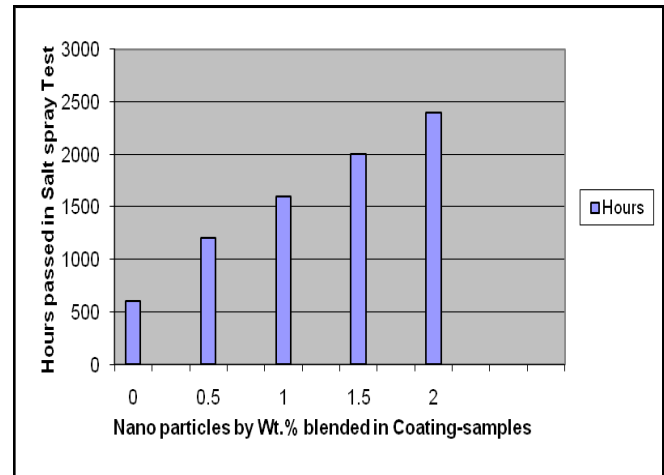


Figure 5. Salt spray test of coated panels having weight % ratio (0, 0.5, 1.0, 1.5, 2.0) of SiO₂ nano pigment particles.

However, by using nanoparticles, scattering of light can be reduced significantly. Nano powders of particle size around 40 to 60 nm are effective fillers. Nanoparticles such as ZnO and SiO₂ have been embedded in ultraviolet (UV) curable lacquers, resulting in improved abrasion resistance. Nanoparticles have been shown to improve the mechanical properties even at low loadings and due to their small particle size; they do not affect the transparency of clear coats. Scratch resistance also improved further due to homogeneous distribution of nano particles in polymers. Even a small amount can retain the appearance of surface without any negative impact on coating and its gloss [10,11,12,25].

Super hydrophobic surface with self cleaning property:

Addition of nanoparticles to coating systems increases its surface area and pore volume, which in turn increases the surface roughness. Increase in roughness increases the contact angle of water and other solvents significantly and hence decreasing the surface tension of coated surfaces.

Hydrophilic Surface: Nano particles may increase surface free energy and thus improve the wetting behavior of water and other solvents. They results in antifog effect due to maximum spreading of water droplets. They also affect capillary flow in micro structural surfaces.

Fire resistant property: Most of the flame retardant coating like ammonium polyphosphate and melamine lose their effectiveness as fire resistant because of their reduced mechanical properties and reduced char formation in fire and hence get easily detached from the substrate. Mechanical and chemical properties of flame retardant coating can be improved by incorporating nano concentrates like nano sized magnesium aluminum –layered double hydroxides (LDH) to different flame retardant coating system. Also, specific amount of nano LDH improve the fire resistant and char formation properties of flame retardant coating. Nano-LDH

absorbs the heat and send out water and carbon dioxide when burns and hence lowers the temperature of substrate along with enhancement in char formation [11,12, 25].

High performance coating: Using nanotechnology, it is possible to create better pigments and coating systems that can give UV alternation, transparency to visible light according to need, along with different colors. Nano-coating can be in great demand for automobile industry. Arrangement of nano pigments can be changed in paints by altering electrical field. Thus, paints can change its color as a function of voltage. This concept can be highly applicable for automobile sector. Nano-sized pigments particle having narrow particle size distribution packed well at the surface of the film resulting in a uniform surface finish. This uniform surface complemented by high scattering power of nanoparticles gives excellent gloss properties to coating systems.

Self-cleaning property: Lotus effect (self cleaning surface of lotus leaves) is very popular in nano-coatings. They check even very fine dirt or droplets from being accumulated on coated surfaces. If nano-coating is applied to glass surface, nanoparticles will interact with ultra violet rays, loose down the dirt particles and than using water, dirt will be distributed across the surface. So, in such glasses dirt can easily be cleaned.

Anti-fouling property: Nano-coatings inhibit the adhesion of microbes and marine fouling organisms. Establishment of nano-structure results in appreciable reduction in interaction between germs and surface. Nano-coating helps in reduction of germs, virus and algae by oligo-dynamic effect of metal component.

Main issues of nano coatings

- a) Main problem in using nano particles for coating purpose is dispersion and stability of nanoparticles. Agglomeration may take place because of high surface energy possessed by nanoparticles due to their large surface area.
- b) Pigments may lose their color on reducing their size to nano level below 20nm and hence will lose their opacity.
- c) Stable binder is required to inhibit photocatalytic activities of nano TiO_2 .
- d) Hardening problems of ultrafine powder.
- e) Surplus use of nanoparticles may confer delivery to unwanted environmental problems, such as newer type of toxic materials and other environmental hazards. Ultrafine particles can catalyze chemical reactions inside body which might be dangerous to human health.

Major benefits of nano-coatings [2, 4, 25]:

- a) Better adhesion on different types of materials
- b) Superior retention of gloss and mechanical properties
- c) Better Scratch resistance.
- d) Anti- reflective in nature
- e) Chromate and lead free
- f) Self-cleaning property: easy to clean surface.
- g) Better surface appearance
- h) Better chemical resistance
- i) Decrease in permeability; resistance to corrosive environment and hence better corrosion protection.
- j) Better thermal and electrical conductivity.
- k) Optical clarity.
- l) Anti-fogging, anti-fouling and anti-graffiti properties.

Outlook of nano coatings

Potential of nano-coating appears very intense. Development in nanotechnology will improve the properties of surface coatings even in next generations day by day. Automobile sector is going to get a lot of benefits from nano materials modified coatings. Objective for future nano-coating would be to formulate such a system in which center will have one set of properties and exterior will have other set of properties.

IV. CONCLUSION

From the time of prehistoric age to modern nano composite coatings, surface coatings have always been pioneer in the areas of surface protection as well as decoration. Whether it might be prehistoric paint or recent paint coatings including nano coatings improve life-span of materials and products by protecting them even in the aggressive environment. The automotive industry is one of the most important consumers of nano technology. Nano coatings knock and solve the several coating-troubles especially for automobile industries. Automobile industry is getting a lot of benefits from nano coatings. Objective for future nano-coating would be to formulate such a coating system in which center will have one set of properties and exterior will have other set of properties for the purpose of better protection and decoration as well.

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