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Environmentally Friendly High Performance Waterborne Coatings

A. Toussaint, M. De Wilde : CoRI , Avenue Pierre Holoffe, 1342 Limelette, Belgium

P. J. Thornton, J. Mulvihill : FORBAIRT, Glasnevin, Dublin 9, Ireland

C. Carr, R. Brown, M. Taylor: PRA, Waldegrave Road 8, TW118LD, Middlesex, Teddington, UK

P. Vink, F. Molenaar : TNO Centre for Coatings Research, Schoemakerstraat 9'7,
P.O. Box 6034, 2600 JA Delft , The Netherlands

B. Kronberg, T. Svanholm : YKI, Drottning Kristinas vag 45, P.O. Box 5607, 11486 Stockholm, Sweden

Abstract

Problems that impair the inroad of latex paints into the industrial sectors are their permeability towards water vapour and other polluting gases (due to improper coalescence and even insufficient sintering of the latex particles] and their inability to impart sufficiently smooth surfaces (due to improper rheology and pigment flocculation) compared to solvent-based paints, resulting for painted substrates in poor resistance to atmospheric and chemical attack and poor aesthetic properties.

Therefore it appears clearly that the formation of pore-free and smooth continuous films from latex paints is of considerable industrial importance and constitutes a prerequisite condition for them gaining share into the industrial market.

The different investigations carried out by the partners during this four-year research project have allowed to make evident and evaluate:

the physico-chemical parameters influencing pigment flocculation, the object being to obtain the best pigment distribution and dispersion;

the physico-chemical parameters influencing the forces acting in promoting or impeding the sintering (and the coalescing) process of the latex particles during film formation.

the factors governing the efficiency of thickeners in controlling the theological properties of the latex-based paints on storage, at application and during drying as well.

1. Introduction

a. Problems related to film formation

Ideally, unpigmented latexes are dispersions of very small and discrete spherical particles of high molecular weight polymers in an outer aqueous phase, stabilised against flocculation by the addition of surfactants and/or protective colloids. The film formation proceeds as follows. Water evaporates from the bulk latex until the particles are packed in the densest ordered array. After that stage, water continues to evaporate from the interstices between the spheres in close contact; the particles may deform into polyeders and coalesce. A continuous and pore-free film can ultimately result, or not, leaving micro pores, depending the challenge between the driving forces which promote coalescence and the forces which impede it. Investigations led the stress on the importance of polymer/water phase interracial tension and capillary forces as driving factors. Recent theoretical considerations make obvious that these forces are complementary, but this needs to be verified thoroughly.
On the other hand forces which impede coalescence are assigned mainly to the mechanical resistance opposed by the latex spheres to deform under the action of the driving forces.

b. Problems related to pigmentation

It is well known that for maximum scattering efficiency (and hiding power), it is necessary that the pigment is distributed evenly as single crystals throughout the film, and that the attainment of good pigment dispersion is also important to achieve good gloss and good barrier properties.
The problem is that only an estimated 20-250/0 of pigment exists as single crystals due to flocculation and purely geometric and statistical reasons. Variables like particle size distribution of the pigment as well as of the latex, particle surface charge density and ionic strength of the medium seem to play an important role in that process, but do not have been interrelated until now.

c. Problems related to application properties

The demands placed on paints are conflicting requirements which can be summarized as follows :
- high viscosity at low shear rates to prevent pigment sedimentation during storage and to avoid sagging; suitable low viscosity at high shear rates to allow easy application together with sufficient film build-up.
- the viscosity, although increasing during drying, must remain low enough the time required to allow any irregularities in the layer to even out (levelling).
This means that the paints need to present shear-thinning characteristics.
It has been experimentally shown that this can be achieved by adding to the paints small amounts of materials, called thickeners, which confer to the paint more or less pronounced pseudo plastic and viscoelastic properties, depending on their nature and amount. This thickening effect results from very complex interactions between the thickener molecules and the other paint constituents, but these interactions are still poorly understood.

d. objectives of the Project.

To palliate all these problems the aims of the proposed research are :

   to make evident and evaluate the physico-chemical parameters influencing pigment flocculation, the object being to obtain the best pigment distribution and dispersion;

   to make evident and evaluate the physico-chemical parameters involved in the coalescing process of the latex particles during film formation.

   to make evident and evaluate the factors governing the efficiency of thickeners in controlling the theological properties of the paints at application as well as during drying.

Combining theoretical principles and the experimentally determined physico-chemical parameters leading to complete coalescence of latex particles and those *governing the* achievement of optimized pigment dispersion and theological properties, the objectives of the research are :

   to develop an explanative model and guidelines to formulate high performance latex paints for use by the industrial collaborators to develop their own proprietary systems;

   to enable the European paint and raw materials industry to share the existing industrial market and to be competitive to others, like Americans and Japanese, who are working extensively to develop such systems.

2 Technical Description and Results

The different investigations carried out by the partners during this four-year research project have allowed to make evident and evaluate:

- the physico-chemical parameters influencing pigment flocculation, the object being to obtain the best pigment distribution and dispersion;
- the physico-chemical parameters influencing the forces acting in promoting or impeding the sintering (and the coalescing) process of the latex particles during film formation.
- the factors governing the efficiency of thickeners in controlling the theological properties of the latex-based paints on storage, at application and during drying as well.

2. 1. It has been shown that the onset and rate of particle sintering can usefully be determined by resistivity and gravimetric measurements combined with non contact profilometry thickness measurements. Although very difficult to assess unambiguously it was found that Atomic Force Microscopy combined with water vapour permeability and water uptake measurements were suitable and most informative techniques in that respect.

2.2, As far as film formation from latexes is concerned *it would probably be advantageous to make use of two different coalescing agents in* thermoplastic latex formulations. *One of them should be hydrophobic* and have a distribution coefficient as low as possible so that it can be uniformly distributed into the particle cores and plasticize them, allowing them to deform and make easier further polymer interdiffusion across the particle boundaries (when coalescence has occurred). The *second coalescing agent* or cosolvent should have a hydrophilic character and *partition preferentially into the hydrophilic* **membranes.** Its role would mainly

be to promote membrane fragmentation or deactivation during the sintering process thus allowing the particles to coalesce. It could be relatively volatile.

This was the role we have attributed to EB in order to explain its "good" coalescing agent behaviour not withstanding its apparent unfavourable distribution coefficient. An underlying idea is that a plasticizer of medium efficiency but mainly located in the membrane may be as efficient as a "good" one uniformly distributed in the particles.

We would like to stress here the fact that _even in the case of soft latexes a coalescing aid would generally be required_ to secure the particles coalescence and their further autohesion. In that case however, a hydrophilic type which will partition in the membrane only could be sufficient resulting in smaller amounts to be used.

2.3. A Solvent Computer Programme, based on thermodynamic properties of the compounds has been extensively used to calculate the evaporation rates of water/solvent mixtures and the composition of the remaining aqueous phase versus drying time. Combined with weight loss measurements it has been shown that the amount of surfactant present in the water phase, the nature and amount of coalescing agent and thickener added, do affect the drying time (evaporation rate) of the latexes, in some cases accelerating it, in others lowering it, depending the system latex/coalescent/thickener/excess surfactant.

2.4. The follow-up of the viscosity changes of a film during drying was shown to be easily recorded by means of the TNO-Filmviscometer. This instrument, developed specifically for this purpose, determines the time $t(\eta_{max})$ to reach the fixation of a *rolling* ball and the viscosity change profile versus drying time. The viscograms so obtained could be correlated to the 'open time' and to levelling/sagging properties. This technique can be and has been extensively used to study the influence of the nature and amount of thickeners and coalescing agents used alone or mixed and the way their interaction influences on these properties.

The instrument measures more than only an equivalent value for the initial in can viscosity as it is able to detect complex rheological changes in the film during its drying and which are governed by physical changes induced by the evaporation process.

It has been shown that the theological parameters in-can viscosity η, the real part of the complex viscosity η' (related to viscous behaviour under dynamic shearing) and the storage shear modulus G (related to the elastic behaviour)of the initial in-can pigmented and unpigmented formulations are in line with the initial total solids volume fraction, and give generally very good agreement with the respective $t(\eta_{max})$ values. The contribution of the polymer particles eventually corrected for swelling by coalescing agents is generally predominant over that of the pigment volume fraction for PVC below ca. 20%.

Generally speaking: _the lower the total volume fraction, the higher $t(\eta_{max})$, and the better flow/levelling and the worse sag_ resistance. The order of $t(\eta_{max})$ is in line with the degree of swelling of the latex particles by the coalescent present.

The theological parameters η, η' and G' agree with respect to levelling/sagging behaviour, as far as these latter are discriminating, since it has been theoretically shown that both viscous and elastic behaviour can reduce the extent of levelling and sagging finally reached. _G' should be as low as possible since it has a retardation effect on levelling._

Thus, the levelling behaviour could be related to initial (dynamic) theological parameters. Spattering has been correlated to the magnitude of the storage modulus G'.

2.5. Dynamic viscosity measurements have confirmed that the associative thickeners (AT) form transient networks in aqueous solutions, showing a strong concentration dependence, but that, in accordance with a model proposed at YKI, these are affected by the presence of surfactants and only to a minor extent by dispersants. The rheological properties of their aqueous solutions seem to be insensitive to the ionic strength of the medium but very sensitive to the addition of coalescing agents.

Also, in particular, care has to be taken when introducing surface active species in a formulation containing AT. Since in general the viscosity and the storage modulus at low shear rates are drastically affected. An increase in η and G' with increasing concentration up to a maximum, which occurs at the same concentration for both curves, is observed, after which η and G' decrease on further addition of surfactant. This effect seems to be less important at high thickener concentration than at low concentration. The effect depends of course on the nature of the thickener.

This behaviour has been ascribed to the formation of mixed micelles that include the hydrophobic parts of both surfactant and thickener. At low to moderate concentrations, the surfactant increases the number of crosslink sites and/or the strength of the crosslinks thus increasing viscosity but at high concentrations the mixed aggregates contain mainly surfactants and the probability of finding many thickener end groups simultaneously within the same micelle decreases. As a consequence, the number of thickener molecules able to form elastically effective crosslinks in the network decreases as does the viscosity.

At the surfactant concentration at which the maximum in viscosity is obtained, the thickener solution is more elastic than a solution of comparable viscosity containing only the thickener.

As a guideline and with the concern _to reduce the risk of obtaining a paint formulation which is inefficiently thickened with an AT due to the heedless introduction of too much surfactant, it should be advantageous to choose an AT characterized by a relatively "high" c_{max}._ In this context c_{max} refers to the maximum of the storage modulus (G') or the low shear viscosity (η_0) of the aqueous AT solution as a function of the molar ratio of the concentration of a relevant surfactant added and the AT. A representative surfactant is e.g. nonyl-phenol- ethoxylated (9 EO) sulphate (Fenopon EP 110).

The thickening effect of AT in latexes has been attributed to their interaction with hydrophobic sites at the latex particle surface. These hydrophobic sites are all potentially able to co-operate in forming a continuous network, acting as junction points in the 3D structure and thus contributing to the increase of η and both the pseudoplastic and pseudoelastic properties of the thickened latex.

As a guideline: at a given volume fraction , latexes with small particle size present of course a larger number of potential crosslink points at short distances from each other and will thus require less thickener to reach the same level of thickening.

However, in the case of latexes with hydrophilic membranes, an alternative mechanism has been suggested, i.e. ion dipole interaction between carboxylate groups extending from the surface of the latex particles and the ether linkage of AT as far as HEUR are concerned.

It has been shown that competitive adsorption of AT on latexes is determined as expected by the surfactant type and amount as well as by the thickener structure. It was found that HEUR thickeners adsorb stronger than HMHEC which in turn adsorb stronger than HEC.

2.6. Pigmentation effect on the state of dispersion, optical and theological properties.

A modelling program has been developed at PRA to simulate the packing of up to three types of spheres in a cubic cell with periodic boundary conditions. The spheres can be of different sizes but in the model each

type was monosized.

Extensive Monte-Carlo rums have been used to approach equilibrium and it is now possible to create log-normal size distributions for investigating the effects of polydispersity assuming random packing as it is the most likely to occur *in* reality.

Many assumptions (including monovalent electrolytes and constant surface potentials) are made in the calculation of the screened electrostatic forces, some of which may not be valid. In addition, the true ionic strength in the systems we are trying to model is uncertain. Nevertheless, the low surface potential of -24 mV assumed for pigment clearly contributes to the close clustering of pigment spheres.

Repulsive potentials due to polymeric ('steric') stabilization are useful from a modelling point of view because they can remove the primary minimum (if this is not too deep) and replace irreversible coagulation by reversible flocculation, thereby validating the use of the Monte Carlo method. The type of steric stabilization in real systems is unlikely to correspond to those for which the theory was developed, but this may not matter too much provided rough estimates of important parameters such as layer thickness are available.

Further complication comes from the repulsive 'structural' or 'hydration' forces due to the hydrophilic nature of the surface of TiO_2. Since the surface of the pigment crystals is likely to be modified the extent of these forces is hard to predict. They are likely to be more important at high ionic strengths but the explicit dependence is not known so they are hard to incorporate into a model if ionic concentration is to be varied.

It is not straightforward to apply results from these models to real paint systems. The earlier hard-sphere models were used to explore geometric packing effects which will probably appear in the final paint. However the models using interparticle potentials can (at present) predict only equilibrium distributions in dilute dispersions, Although this may represent the situation in a paint tin before application, the high shear forces during application are likely to break Up loose floes and remix the particles. If the paint dries slowly it is possible that clustering could reestablish themselves. Whether the models have any useful relation to real systems can only be discovered by experiment.

The main predictions obtained from the model are:

- Single Crystal Fraction decreases exponentially with PVC for a given pigment/latex size ratio
- Effect of PVC follows a simple mathematical relationship based purely on geometric factors.
- Single Crystal Fraction increases as latex particle size decreases.
- Increasing ionic strength first causes clustering of pigment particles. At very high ionic strength clustering of latex particles is predicted.

In a real pigmented latex formulation, distribution of pigment particles will be influenced by many chemical and physical factors in addition to the geometric effects. These include: the initial state of dispersion of pigment, the surface potential of pigment and latex, the ionic strength of medium and the viscosity.

Screening experiments were designed to identify which factors are important and to find out whether suspected interactions between parameters had a marked effect on the optical, barrier and theological properties in real formulations. The variables considered were : pigment grade, PVC, dispersing agent and its concentration, thickener and its concentration, mixing technique, latex particle size and pH.

Only *four variables appeared to be significant. The* most important is *the pigment grade.* The reason why

pigment grade affects the quality of the film can be explained from the different interactions which will arise in the wet state and during drying.

The incorporation of *thickener* into the formulation in most cases *retarded the optical properties of the film*. *This* is probably because of the associative nature of thickeners causing pigment particles to interact and to flocculate. *The effect of thickener was influenced by pH*, HEC being better at high pH, while ADX 439 appears to perform better at lower pi-I.

The final term demonstrated to be significant is the interaction between *PVC* and dispersing agent concentration. The higher the PVC the higher the dispersant concentration needs to be, the converse also being true. This is not wholly surprising as every pigment-dispersant combination has an optimum concentration of dispersant (w/w pigment) for maintaining dispersion.

The latex particle size falls just below the cut off point for significant variables although it is expected that latex particle size will affect the properties of the film. It is also interesting to note that properties of the film were better with the large particle size latex. This is contrary to what is suggested by work on model systems to date. The explanation is probably the competitive nature of interactions.

The mixing technique used for letting down the pigment millbase appeared particularly insignificant.

Statistical analysis of the experimental results (single crystal fraction, gloss, flocculation gradient, application properties) has led to the following conclusions. The dominant factors affecting pigment dispersion (single crystal fraction) are pigment grade and PVC,

- Composition of surface treatment alone cannot be used to characterise the pigment grade. The morphology or structure is important.

- Thickener type is the next most important, but the most effective type is probably specific to the pigment grade and dispersant type used,

- An optimum latex size between 100-200nm and an optimum pi-i of ca 9 is suggested for the systems studied.

- The optimum parameters are probably specific to individual pigment grades.

Variables which strongly influence SCF would also influence gloss, flocculation gradient and opacity and *other* rheological properties. Experiments have led to the following:

Small latex particles improve gloss and flocculation gradient in accordance with prediction from hard sphere model systems. It should also be noted however, that the screening design suggests that very small particle sizes (~90 nm) are detrimental. This data may indicate the presence of an optimum latex particle size between 100 and 200 nm but there is insufficient data to prove this and further investigation is required.

In conclusion, optical properties are dominated by PVC. Latex particle size is also a strong influence with small sizes {down to 185nm) giving best results. Other factors which are possibly important are thickener concentration and pH.

Sag and levelling are determined primarily by thickener type and concentration as would be expected. Coatex appears to be the most effective thickener compared to ADX. It gives better sag resistance and improved opacity with RCL-6 and RCL-628.

High thickener concentrations improve sag resistance and impair levelling as would be expected.

Sag resistance is improved but levelling worsened with small latex particles. This is probably a consequence

of improved thickening at low particle sizes. A supposition which is reinforced by a strong interaction term between thickener concentration and par-tide size. The direction of the effect points towards synergy at small size and low concentration or large size and high concentration. Also, an improvement in sag resistance was observed with small particle size latices with high thickener concentrations. For levelling, antagonism occurred between particle size and thickener concentration. Levelling was particularity poor at low particle size with high thickener concentration.

Dispersant type appears as a primary variable affecting sag resistance in RCL-6 but also shows a strong interaction term with thickener type in influencing sag and levelling. Synergy was observed between Coatex and Tamol. The effect of dispersants on the action of thickeners is well recognised. The results would also imply that the chemical interaction between Tamol and thickener observed at YKI is beneficial to sag resistance.

The other important. variable was ph. High pH promotes good sag resistance while low pH promotes good levelling. This is probably due to competitive interactions (pigment/latex/dispersant/ thickener) being pH sensitive.

It has been shown that sag and levelling are dominated by thickener type and concentration as expected but pH and dispersant type and concentration need also to be considered.
However the optimum parameters are probably specific to individual pigment grades remembering that the screening design showed that the most important variable affecting pigment dispersion is the pigment grade itself.

2.7. It has been shown that the dynamic mechanical characteristics of latex films, plasticized or not, which were obtained by Dynamic Mechanical Analysis fitted well to a "limited parabolic" theological model instead of to the more commonly used ones (Maxwell, Voigt, Zener, Burgess). The results showed that the WLF concept was applicable so that master curves for storage and loss moduli could be drawn and the relaxation times of the polymer determined.
From this model a new definition has been given for the minimum film formation temperature (MFFT) and a mechanical description of the particles sintering process is proposed taking into account the forces promoting the particle deformation and the viscoelastic properties of the particles which are opposing, using a 'limited biparabolic' rheological model. From the resulting mathematical model and from a practician or formulator point of view it was derived that *the relaxation time τ of the polymer constituting the particles must be lower or equal to a. certain value which can be calculated for full sintering to be achieved* at the time - not before, nor later- the aqueous phase has completely left the film under the given drying conditions. This value of the relaxation time depends on the magnitude of the driving forces for sintering, the viscoelastic properties of the particles' constituting material (eventually plasticized), the particle radius and the drying time. All these parameters can be calculated or determined experimentally. The required value of the relaxation time implies also that for the *given* set of film drying conditions, the particles material must present a glass transition temperature Tg equal or lower than a certain calculable value (WLF or Adam-Gibbs relationship].
Alternatively and as a guideline this also means that under given drying conditions a void-flee film will only

be obtained at a temperature (or higher) for which the relaxation time of the latex polymer has the required value and that temperature will be called the MFFT. It follows from this new definition that *for a given latex the MFFT is dependent on particle size, the drying time, i.e. the thickness of the film and the evaporation rate of the aqueous phase (RH, air circulation) and on the nature and intensity of the driving forces, who are due to capillary or osmotic forces (open time) or both.*

It must be stressed here that though Tg and MFFT are closely related, their meaning is quite different and that they may not be used indifferently from each other.

2.8. A model is proposed allowing the *a priori* calculation of the lowering AT of the glass transition temperature {or equivalently of the MFFT) on the addition of a given amount of coalescent or inversely of the volume or weight fraction of coalescing agent to be added to a latex in order to obtain a given lowering AT of its MFFT. It is based on the flee volume concept extensively used in the polymer field and uses physical parameters that can either be found in the literature or easily experimentally determined. It has also been verified to be applicable to mixtures of coalescing agents.

A thermodynamically derived relationship has been proposed allowing the calculation of the partition coefficients of coalescing agents between the aqueous phase and the latex particles. It is based on the chemical potential theory and makes use of the very recent Mobile Order Theory developed by Huyskens in 1985.

3. Conclusions

The different investigations carried out by the Consortium during this four-year research project have allowed to make evident and evaluate:

the physico-chemical parameters influencing pigment flocculation the object being to obtain the best pigment distribution and dispersion;

- the physico-chemical parameters influencing the forces acting in promoting or impeding the sintering and the coalescing process of the latex particles during film formation.

the factors governing the efficiency of thickeners in controlling the theological properties of the latex-based paints on storage, at application as well as during drying.

More particularly, from the model set up for the pigment dispersion study it is predicted that :
- Single Crystal Fraction decreases exponentially with PVC for a given pigment/latex size ratio
- Effect of PVC follows a simple mathematical relationship based purely on geometric factors.
- Single Crystal Fraction increases as latex particle size decreases.
- Increasing ionic strength first causes clustering of pigment particles. At very high ionic strength clustering of latex particles is predicted.

As far as real pigmented systems are concerned it was shown that:
- pigment grade and PVC are the dominant factors affecting pigment dispersion.
- Composition of surface treatment alone cannot be used to characterise the pigment grade. The morphology

or structure is also important.

- Thickener type is the next most important factor, but the most effective type is probably specific to the pigment grade and dispersant type used as well.
- An optimum latex size between 100-200nm and an optimum pH of ca 9 is suggested for the systems studied.
- The optimum parameters are probably specific to individual pigment grades.

A Solvent Computer Programme, based on thermodynamic properties of the compounds has been extensively used to calculate the evaporation rates of water/solvent mixtures and the composition of the remaining aqueous phase versus drying time. Combined with weight loss measurements, it has been shown that the amount of surfactant present in the water phase, the nature and amount of coalescing agent and thickener added, do affect the drying time (evaporation rate) of the latexes, in some cases accelerating it, in others lowering it, depending the system latex/coalescent/thickener/excess surfactant.

The follow-up of the viscosity changes of a film during drying was shown to be easily recorded by means of the TN O-Filmviscometer. This instrument, developed specifically for this purpose, determines the time $t(\eta_{max})$ to reach the fixation of a rolling ball and the viscosity change profile versus drying time. The viscograms so obtained are correlated to the 'open time' and to levelling/sagging properties. This technique has been used to investigate the influence of the nature and amount of thickeners and coalescing agents on these properties and on $t(\eta_{max})$.

Dynamic viscosity measurements have confined that the associative thickeners (AT) form transient networks in aqueous solutions but that, in accordance with a proposed model, these are affected by the presence of surfactants and only to a minor extent by dispersants.
It has been shown that in general thickeners did not adsorb onto TiO_2 but did on latex particles depending the extent of their surface coverage by surfactant.
Coalescent did not seem to influence the adsorption of the thickeners. Also, the ionic strength of the aqueous phase did not seem to affect the AT network.
The adsorption of associative thickeners on latexes and hence the drastic viscosity increase at low shear rate is determined by the surfactant amount and type as well as the thickener chemical structure

A model is proposed allowing the *a priori* calculation of the lowering AT of the glass transition temperature (or equivalently of the MFFT) of the particle constituting polymer on the addition of a given amount of coalescent or inversely of the volume or weight fraction of coalescing agent to be added to a latex in order to obtain a given lowering AT of its MFFT.

A thermodynamically derived relationship has been proposed allowing the calculation of the partition coefficients of coalescing agents between the aqueous phase and the latex particles. It is based on the chemical potential theory and makes use of the very recent thermodynamic Mobile Order Theory.

Finally it has been shown that the dynamic mechanical characteristics of latex films, plasticized or not, which were obtained by Dynamic Mechanical Analysis fitted well to a "limited parabolic" rheological model instead

of to the more commonly used ones (Maxwell, Voigt, Zener, Burgess). The results showed that the WLF concept was applicable so that master curves for storage and loss moduli could be drawn and the relaxation times of the polymer determined.

From this model a new definition has been given for the minimum film formation temperature (MFFT). It is thus theoretically shown that the MFFT depends not only on the latex particle size and the modulus of the polymer but also on the drying conditions (temperature, relative humidity, air velocity above the film)

The model allows also the calculation of the viscoelastic characteristics a polymer should present for complete sintering to occur under defined drying conditions.

A. TOUSSAINT

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