ResonantAcoustic® Mixing – Processing and Formulation Challenges for Cost Effective Manufacturing

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Abstract

Sonic mixing technology was developed in the 20th century in the chemical and pharmaceutical industry fields (mixture of powders and fluids). It knows a strong renewed interest in the USA in the early 2000s under the active leadership of the US Department of Energy (Office of Industrial Technology) due to, on the one hand, its high potential in terms of energy bill reducing (by decreasing the production cycle time) and, on the other hand, the flexibility of the technology and its expansion capacity for a wide range of industries.

Through its quality policy of improved commercial, technical, industrial and economic performances, Roxel, European leader in tactical propulsion systems, has invested in a new process to manufacture inert and energetic materials: the ResonantAcoustic® Mixing (RAM) by first purchasing a 1 pint capacity mixer called LabRAM.

Contrary to conventional mixing methods such as impeller agitation found in a planetary mixer, the RAM technology applies low-frequency, high-intensity acoustic energy to create a uniform shear field throughout the entire mixing vessel thus enabling a better quality of the final mixtures in a reduced amount of time.

A wide range of inert materials developed in Roxel were successfully manufactured on the LabRAM such as:

- Epoxy, silicone and polyester inhibitors
- Highly loaded thermal insulators
- Polyurethane liners
- Powders (inert simulant of ignition powder)…

All those materials were characterized in order to verify the conformity of their mechanical, rheological, topographical and thermal properties. The results obtained prove the huge potential that it represents: not only the quality and reliability of the mixture is enhanced but the time of implementation of the mixing cycle is drastically reduced (hours → minutes). Due to the requirements, only the results obtained on an epoxy inhibitor will be presented in this paper.

Such benefits could be for Roxel an additional advantage in maintaining its leadership and in gaining access to new markets in a highly competitive sector. Those benefits will be discussed through Roxel presentation.

Keywords: ResonantAcoustic® Mixing • Acoustic energy • Resonance frequency • Bulk mixing • Micro-mixing • Mechanical, topographical and thermal properties • Coefficient of variation.

1. Introduction

The ResonantAcoustic® Mixing (RAM) technology is distinctly different than conventional mixing methods such as impeller agitation found in a planetary or speed mixer for example. Conventional methods work by producing bulk fluid flow in a mixture whereas RAM brings to you a new paradigm in mixing that is based on using acoustic energy to create flow in liquids, slurries and powders. So we ought to wonder the consequences that will create such a way of mixing on the materials. The main purpose of this paper is to analyze the effects of RAM

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mixing on the final aspect of an epoxy inhibitor by focussing on its mechanical, topographical and thermal properties. Some advantages and benefits of this technology are also described in various experiments corroborating our results.

2. Equipment and methods

2.1 The used devices

The LabRAM shown in Fig. 1 is a lab mixer with a nominal capacity of 1-pint that is based on the ResonantAcoustic® Mixing technology. At the heart of the LabRAM mixer is a resonant mechanical system, called resonator, which applies high-intensity, low-frequency acoustic energy to create a uniform shear field throughout the entire mixing vessel [1].

Figure 1. The LabRAM mixer
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This mixer can be decomposed in three major components: the resonator, the vibrating plate and the vessel with its vessel holder (see Fig. 2). The mixing process is based on the transmission of an acceleration (measured in units of G’s) to bulk products, from a given intensity of acoustic energy and a specific resonance frequency, to obtain a final homogeneous mixture.

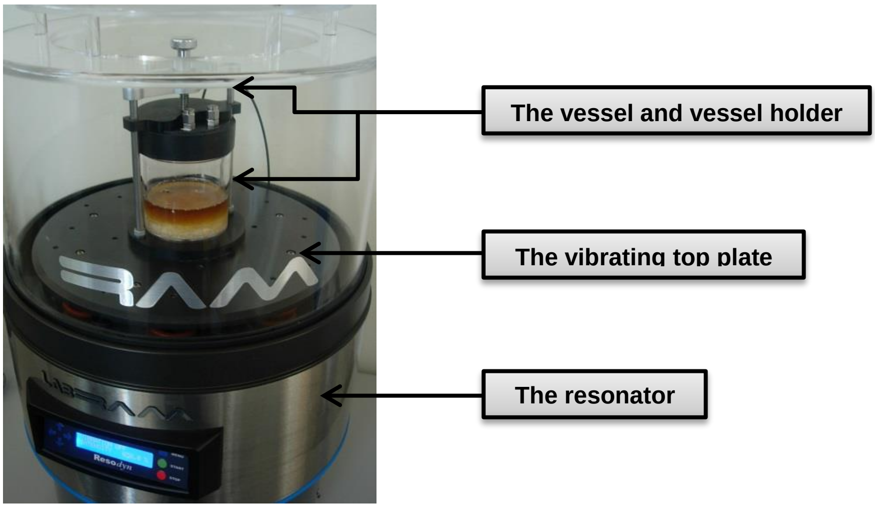


Figure 2. A diagram showing the main components of the LabRAM mixer

The studied epoxy inhibitor was characterized mechanically with an INSTRON tensile machine, topographically with a BRUKER Nanoscope V MultiMode 8 and thermally on the NETZSCH dilatometer DIL 402 C.

2.2 The process

Mixing

All the main steps allowing the manufacture of the epoxy inhibitor with the LabRAM mixer are listed in the Tab. 1.

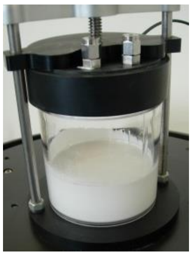
LabRAM trials			
	Trial 1	Trial 2	Trial 3
Order of insertion into the vessel	-titanium oxide powder -hardener (heated at 50 °C) -epoxy resin 		
Quantity used	180 g		
Vacuum	26 inHg	27 inHg	26 inHg
Initial temperature	25 °C	24 °C	24 °C
Mixing cycle	Frequency : 60.19 Hz Prior degassing for 2 min Proceed by stages : - 30 s at 10 % intensity - 1 min 30 s at 15 % intensity - 3 min at 30 % intensity - 1 min at 45 % intensity Acceleration transmitted to the mixing : ~ 25 G's 		
Final temperature	29 °C	27 °C	28 °C

Table 1. A table presenting the main steps to obtain the epoxy inhibitor with the LabRAM mixer

Characterization

After the mixing was done, the epoxy inhibitor was casted and cured at 36 ± 3 °C during 18 to 26 hours.

Tensile tests:

The operating mode was conducted as followed:
- -tensile specimen of type AFNOR H2;
- -ageing of the sample: 10 ± 2 days at 20 °C;
- -tensile test temperature: 20 ± 2 °C;
- -tensile test hygrometry: 35 %;
- -tensile speed of 50 mm/min.

A density measurement was made on one of the tree samples coming from each trial.

Dilatometry:

The dilatometer was operated and programmed according to the following steps:
- -inert gas: N_2;
- -80 °C during 15 min;
- -descent to -70 °C at 2 °C/min;
- -bearing at -70 °C for 15 min;
- -rise to 80 °C at 2 °C/min;
- -bearing at 80 °C during 5 min.

Atomic Force Microscopy (AFM):

The analysis was performed with a PeakForce QNM ("Quantitative Nanomechanical Property Mapping") imaging mode. The samples were prepared with an ultra cryo-microtome system (glass/diamond knives) and the analysed samples were taken from the middle of the plate, in the center of the plate thickness.

3. Results

3.1 Mechanical properties

The study was only focused on the analysis of three main mechanical properties: Young's modulus E, the ultimate tensile strength Sm and the elongation at fracture e_r. A test to calculate the density was made in order to check the integrity of the internal structure (absence of porosity). The obtained results are compared to the specification of acceptance of this epoxy inhibitor and reported in the Tab. 2, 3 and 4.

Tensile specimen	E (MPa)	Sm (MPa)	e_r (%)	Density
1	14.85	6.27	114.71	1.141
2	15.61	6.97	123.91	
3	16.26	6.68	116.10	
4	15.68	6.86	122.40	
Specification of acceptance	$5 \leq E \leq 50$	$3 \leq Sm \leq 15$	$50 \leq e_r \leq 200$	$d_{th}=1.14$

Average	15.60	6.70	119.28
Extent	1.41	0.70	9.20
Standard deviation	0.58	0.31	4.55
Variance	0.33	0.09	20.72
Coefficient of variation Cv	0.037	0.046	0.038

Table 2. Mechanical properties for trial 1

Tensile specimen	E (MPa)	Sm (MPa)	e_r (%)	Density
1	20.10	5.47	94.32	1.142
2	18.02	6.12	104.40	
3	20.63	5.41	90.33	
4	17.48	6.07	101.93	
Specification of acceptance	$5 \leq E \leq 50$	$3 \leq Sm \leq 15$	$50 \leq e_r \leq 200$	$d_{th}=1.14$

Average	19.06	5.77	97.74
Extent	3.15	0.71	14.07
Standard deviation	1.54	0.38	6.54
Variance	2.37	0.14	42.84
Coefficient of variation Cv	0.081	0.066	0.067

Table 3. Mechanical properties for trial 2

Tensile specimen	E (MPa)	Sm (MPa)	e_r (%)	Density
1	15.78	5.99	111.47	1.142
2	16.51	6.45	112.27	
3	16.14	6.50	115.94	
4	15.15	6.11	111.10	
Specification of acceptance	$5 \leq E \leq 50$	$3 \leq Sm \leq 15$	$50 \leq e_r \leq 200$	$d_{th}=1.14$

Average	15.89	6.26	112.69
Extent	1.36	0.51	4.84
Standard deviation	0.58	0.25	2.22
Variance	0.34	0.06	4.91
Coefficient of variation Cv	0.036	0.040	0.020

Table 4. Mechanical properties for trial 3

3.2 Topographical properties

Two epoxy inhibitor samples, one manufactured with the LabRAM and the other with a traditional mechanical mixer, were prepared and analysed by AFM PeakForce QNM in order to obtain a mapping of the topographical, mechanical and adhesive surface properties while minimizing the contact force between the tip and the sample (see Fig. 3, 4 and 5).

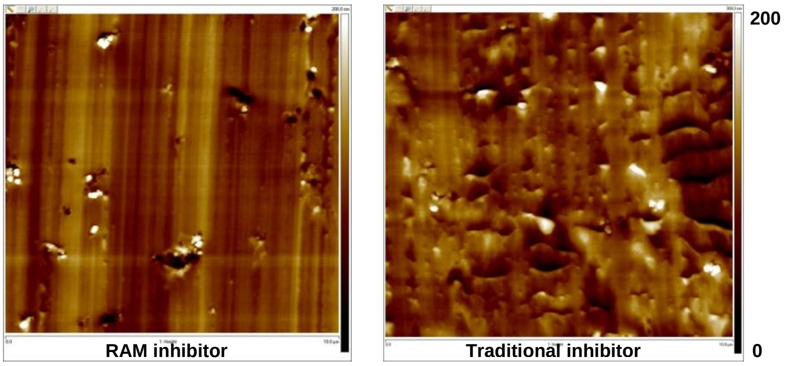


Figure 3. Roughness images of epoxy inhibitor by AFM, 10 µm x 10 µm (scale: 200 nm)

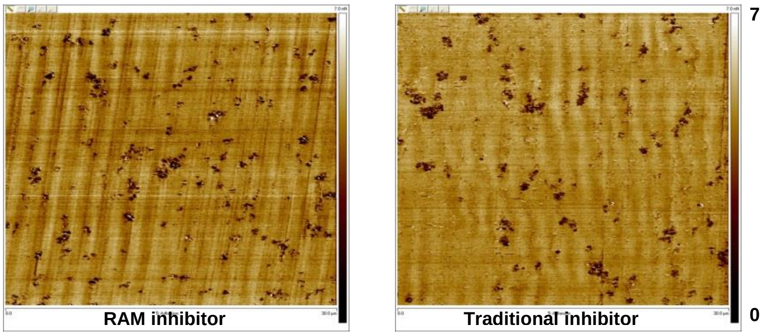


Figure 4. Adhesion images of epoxy inhibitor by AFM, 30 µm x 30 µm (scale: 7 nN)

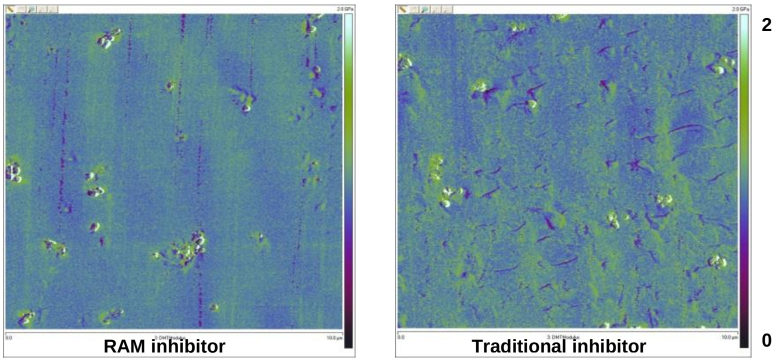


Figure 5. Modulus images of epoxy inhibitor by AFM, 10 µm x 10 µm (scale: 2 GPa)

3.3 Thermal properties

Three measures of the linear expansion coefficient of the epoxy inhibitor were made on two samples coming from trials 2 and 3. They were measured on a temperature range of +60°C to -40°C in descent and -40°C to +60°C in rising. The obtained results extracted from heat diagrams are listed in Tab. 5.

Samples	**Measures**	**Average linear expansion coefficient in descent of temperature Range : +60°C to -40°C (ppm.K^{-1})**	**Average linear expansion coefficient in rising of temperature Range : -40°C to +60°C (ppm.K^{-1})**
Trial 2	1	166	147
	2	166	143
	3	165	140
	Average	166	143
Trial 3	1	153	139
	2	162	140
	3	164	145
	Average	160	141
Total average		**152.5**	

Table 5. Linear expansion coefficient of the epoxy inhibitor

4. Discussion

From the data obtained during the tensile test, it is obvious that the mechanical properties are in the specifications for acceptance of the epoxy inhibitor, so this new device can definitely be used to elaborate this material. A statistical analysis has been made on the relative standard deviation (RSD) of two of the three main studied mechanical properties, the ultimate tensile strength and the elongation at fracture, in Fig. 6 and 7. Indeed, we deliberately choose to not proceed on the analysis of the Young’s modulus insofar as the inhibitor has a glass transition temperature that is very close to the characterization temperature which could lead to important dispersions. This analysis compares the RSD obtained using the LabRAM process and the one using a conventional process meaning the mixing with impellers.

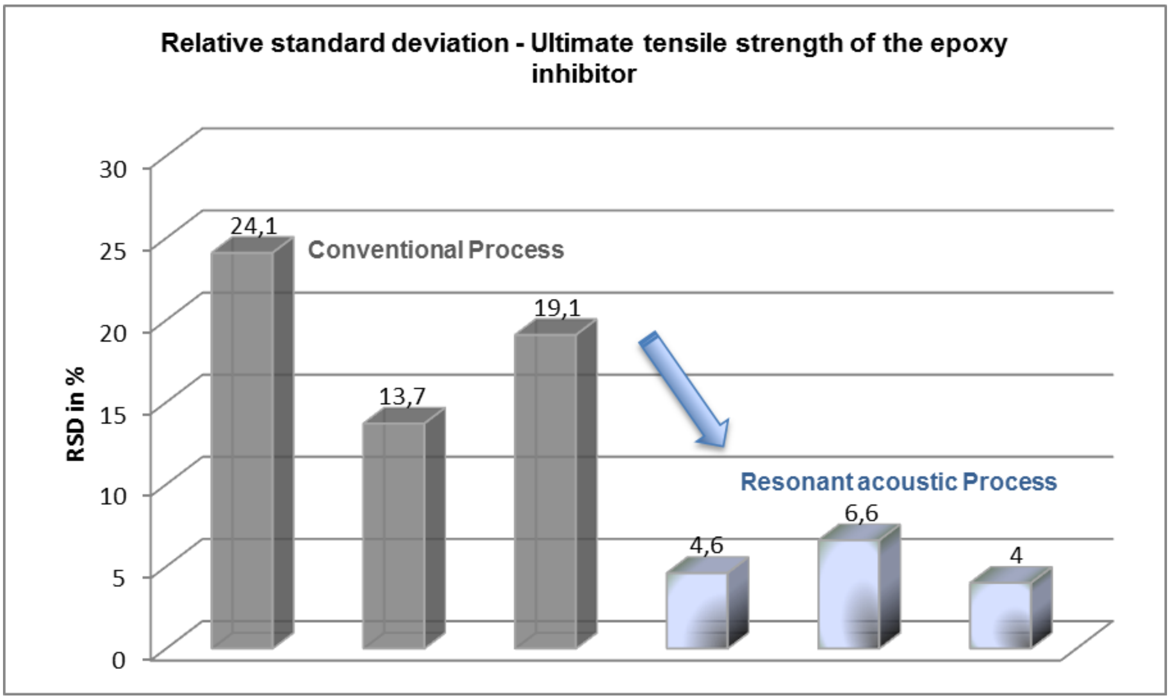


Figure 6. Comparative analysis of the RSD on the ultimate tensile strength according to the mixing device

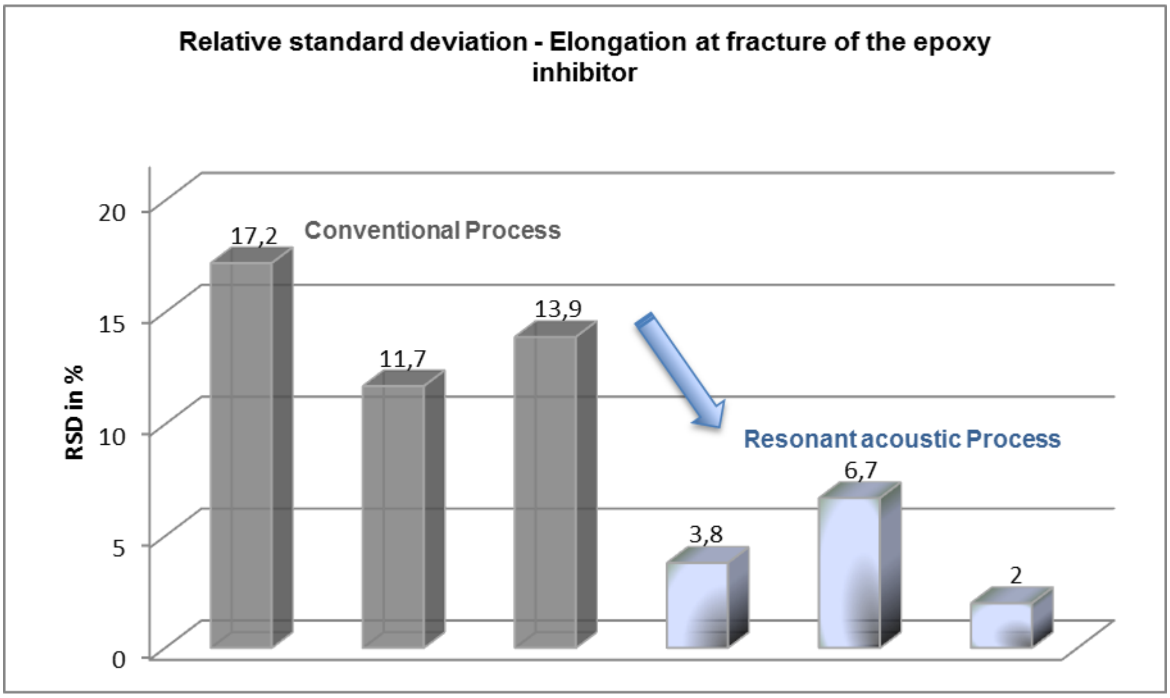


Figure 7. Comparative analysis of the RSD on the elongation at fracture according to the mixing device

The graphics shows the good reproducibility and reliability (reduction of the RSD) of the RAM technology. Also, with regard to the density, the values given, to compare with the theoretical value of 1.14, confirm the quality of the final mixture. Moreover if we take as a reference the expansion coefficient of the epoxy inhibitor obtained with a conventional mixer: $\alpha \approx 150.5$ ppm.K^{-1}, we get similar results with the one manufactured with the LabRAM mixer.
Fernando Muzzio, a Professor at Rutgers University, who is currently conducting comparative studies between conventional mixing technologies ("double cone" blender) and RAM technology (LabRAM) for the implementation of powder compositions [2], did an analysis based on a statistical evaluation (calculation of the relative standard deviation) on the measured performances of each technology and came to the conclusion that although mixing times are greatly reduced when using RAM technology, the quality of the final mixture is improved which corroborates our results.
Furthermore, Michael Mangum, a Senior Research Chemist at Goodrich Company, is in charge of development activities on energetic materials. One of his goals is not only to find a way to reduce manufacturing cycle time but also to ensure a better homogeneity of the product and thereby to reduce the dispersions on the performances [3]. Therefore, he also did a statistic analysis on the coefficient of variation (Cv) calculated from calorimetric potential measurements on powder mixtures. His results also show that the Cv is tremendously reduced when using a LabRAM mixer.

Regarding the AFM PeakForce analysis, on the micron or sub-micron observed scale, the main difference we notice between the RAM and traditional epoxy inhibitor is the surface topography, which is significantly more rugged on the inhibitor developed with a classical mechanical mixer (surface roughness is twice as high), with a noticeably poorer filler dispersion than the one obtained with the RAM process. No notable differences were observed in terms of the mechanical properties at this scale, except for the higher scattering of the measured values on the traditional inhibitor, which makes sense given its more rugged surface topography. Those results further confirm the improvement of the quality of the mixture obtained with the RAM technology.

According to Peter Lucon, Director of Technology and Processing at Resodyn Corporation who is in charge of the modelling process activities developed in the company and more precisely on the RAM mixers [4], the ResonantAcoustic® Mixing presents many benefits and features that cannot be obtained on conventional mixers. For example, the acoustic energy fluidizes and mixes the entire reactor contents without creating any dead zones contrary to impeller mixers. He represented the mixing model of the LabRAM as a mass-spring damper system, as shown in Fig. 8.

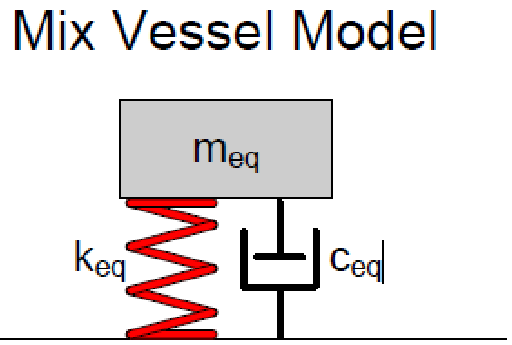


Figure 8. Simplified dynamic model for the mix material in the mixing vessel
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From the differential equation governing the mass movement we obtain the followed equation:

$$Meq\ddot{x} + Ceq\dot{x} + Keqx = f(t)$$

M_{eq}: equivalent mass (constant)
C_{eq}: equivalent damping coefficient (mixing)
K_{eq}: equivalent spring rate (coupling)
f(t): forcing function

The principle of RAM mixers is illustrated in both Fig. 8 and 9.

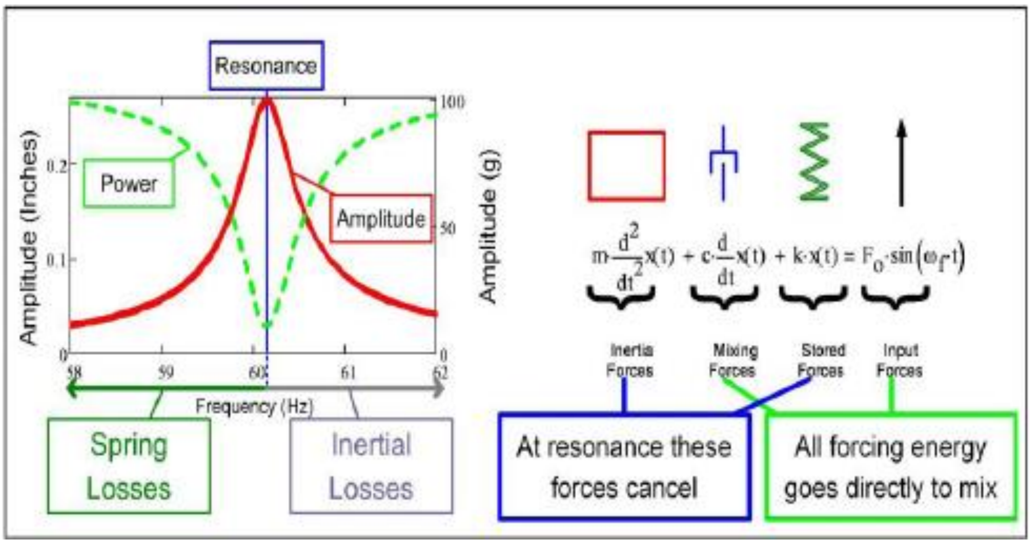

Figure 9. Differential equation and plots of oscillation amplitude and power vs. frequency showing the benefit of resonance
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This system creates during a mixing two main phenomena: the bulk mixing induced by the acoustic streaming (macro-mixing) and the micro-mixing induced by the acoustic field. In doing so, RAM process creates a uniform shear field throughout the entire mixing vessel (see Fig. 10-2) rather than located at the ends of the impeller blades (see Fig. 10-1).

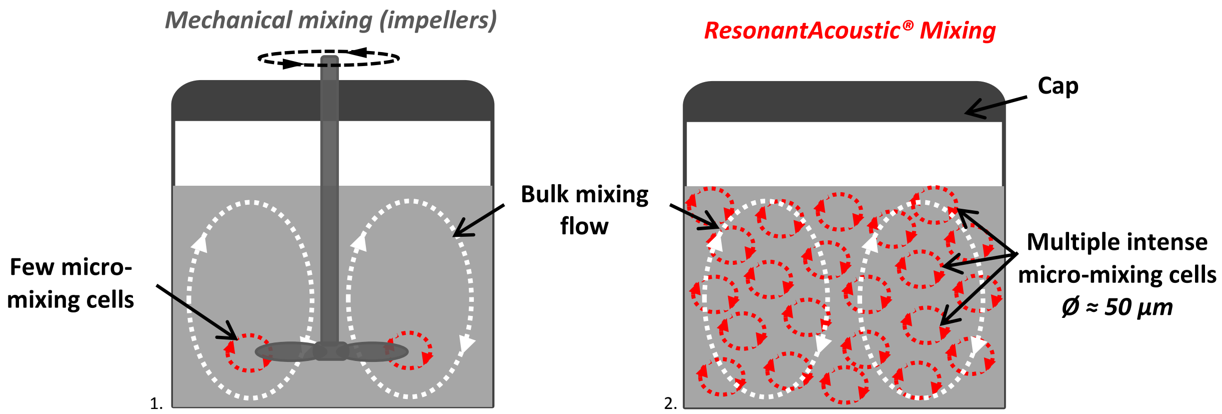

Figure 10. Mechanical mixing vs. ResonantAcoustic® Mixing

It is the combination of all those criteria and more that determine the excellence of the results achieved and efficiency of this invention.

This new technology is still in constant progress and many people try to understand what really happens inside during the acoustic mixing.
However difficult it is to understand the theory of RAM technology, all the results prove the huge potential that it represents. Not only the quality and reliability of the mixture is enhanced but the time of implementation of the mixing cycle is drastically reduced. Indeed, the conventional process requires approximately 4 hours to do the premix and more than 11 hours to obtain the final mixing of this epoxy inhibitor; meaning a total of more or less 15 hours. Compared to the 6 minutes on the LabRAM, the decrease of time allowing a better flexibility in the organization of process operations in workshops and the reduced costs of energy are substantial and significant benefits.

5. Conclusion

The ResoantAcoustic® Mixing technology allows to obtain a better quality of the final mixtures in a reduced amount of time. Impacts on the mechanical, topographical and thermal properties of an epoxy inhibitor have been studied and the results show a significant decrease of the relative standard deviation of the ultimate tensile strength and the elongation at fracture when using the LabRAM mixer. Calculations of the density and expansion coefficient of this material tend to lead to the same conclusion, meaning an improved quality of the obtained product.

Some exciting work is also being performed on propellant while using this vanguard technology and very promising results are obtained.

In practical terms, the use of this new device is quite easy and the results clearly pinpoint the advantages and benefits of this technology but the understanding of the theory explaining why we get such results is still under study. Given the pioneering nature and complexity of this mixer, the precise scientific and technical knowledge will be difficult to obtain on this day.

Some interesting perspectives would be a better insight into the internal mechanisms governing this technology and understanding of the effects of acoustic waves on the mixture. The study on a higher scale (RAM5: 5 Gallons mixer) can also open a large research field.

Acknowledgements

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