

# Is ResonantAcoustic Mixing® (RAM) a Game Changer for Manufacturing Solid Composite Rocket Propellants?

Christopher J. Wright<sup>a</sup>, Peter J. Wilkinson<sup>b\*</sup>, Sally E. Gaulter<sup>b</sup>, Donald Fossey<sup>c</sup>,  
Andrew O. Burn<sup>d</sup>, and Philip P. Gill<sup>e</sup>

**Abstract:** This study is a structured literature review of published ResonantAcoustic® Mixing (RAM) literature, considering the benefits and constraints of using RAM. Focussing on how this will affect the future production of rubbery composite rocket propellants. The main benefits of RAM were found to be shorter mixing time, versatility of mixing and ability to mix higher viscosities than conventional mixers. Facilitating the next generation of composite propellants with improved performance and mechanical properties. Mixed in-situ RAM overcomes viscosity limitations by removing the casting process and has safety and environmental benefits, but does need to be tested at larger production scales. The implications of RAM production on the energetics qualification process was considered. A new framework was discussed based on understanding the entire product development process including ingredient properties, manufacturing processes, and linking this to product performance; through adoption of a digital twin approach with in-situ monitoring. Future R&D focuses on process and material control through a validated model of the mixing mechanisms, linked to material properties and output performance. Validation with scaled up comparative studies and continuous in-situ monitoring. A full list is provided in the conclusions. Overall RAM offers numerous benefits to mixing existing and new materials with large savings in time, cost, improved safety and is more environmentally friendly.

**Keywords:** ResonantAcoustic® Mixing, RAM, Solid composite rocket propellants, Smart qualification

## 1 Introduction

Mixing is important in both industry and domestically. This could be for mixing food, such as for baking a cake or for mixing medicines using micro or nano-scale ingredients. The chemical Industry has developed much of the theory of mixing, but many other sectors including food, battery and pharmaceutical all incorporate large scale mixing into their manufacturing processes. Manufacturing made up 15.5 % of global Gross Domestic Product (GDP) in 2017 [1] and in the UK the manufacturing industry accounts for 69 % of research and development expenditure [2].

In the 21st century the world has moved into the Industrial Revolution 4.0, where the Internet of Things, Data and Services connections means that the internet connects each stage of the development, manufacturer and customer focussed parts of a business. This availability of information in real-time enables a new level of control and agility over a product at any stage of its lifecycle [3]. In all industries, this additional information will need to be understood to make the

system or process successful. Mixing is a critical part of the scale-up from laboratory testing, and if the product fails to meet the required product yield, quality, or physical attributes then this could increase the cost of manufacturing significantly or marketing of the product could be delayed due to the cost and time to correct the mixing problem [4]. In 1989, USA industrialists and academics estimated that the cost due to inadequate understanding of mixing, was up to \$10 Billion per annum [5]. This was primarily due to misunderstanding the multi-faceted mixing mechanisms and their impact on end product quality. In particular, the rheological complexity of high viscosity was found to be difficult to scale-up correctly.

The dispersion of fine particles in liquids is a problem for rocket propellants but is also applicable to the chemical, construction and pharmaceutical industries. The fine particles are considered to have at least one dimension with a length between 1  $\mu\text{m}$  and 1 nm [6]. Although this small size is currently aspirational for propellants; 4-400  $\mu\text{m}$  particles are currently used. The smaller particle sizes increase the overall surface area of the material. This will increase the interparticle forces and will form aggregates and agglomerates.

The two stages of viscous mixing are described as:

- (i) Dispersive mixing, where the agglomerates are broken up and
- (ii) Distributive mixing, where the spatial uniformity of all the components is optimized.

Intensive dispersive mixing is needed to break up the agglomerations present while at the same time extensive distributive mixing is required to disperse them into the liquid/continuous medium [4]. The viscosity of the liquid will affect this rate of wetting and the depth of penetration of the solid particles. Other

[a] Ministry Of Defence, Royal Navy, UK

[b] Centre for Defence Chemistry, Cranfield University, Defence Academy of the United Kingdom, Shrivenham, SN6 8LA, UK \*E-mail: [p.wilkinson@cranfield.ac.uk](mailto:p.wilkinson@cranfield.ac.uk)

[c] The Falcon Project Ltd, Westcott, Aylesbury HP18 0XB, UK

[d] BAE Land UK, Glascoed, Usk, NP15 1XL, UK

[e] ROXEL (UK Rocket Motors) Ltd, Summerfield Lane, DY117RZ, UK

factors also play an important role in this, such as entrapped air, the wetting of the solid and the porosity of the solid [7,8]. With higher viscosities an increase in mixing time or additional of mechanical mixing is needed. High shear devices such as mills or paddle/bladed mixers can fragment the agglomerates. While milling is used for mixing energetics such as pyrotechnics and gunpowder the intense milling action will increase the temperature of the mixture and there is a risk of incident for solid-liquid mixtures.

Figure 1 shows the viscosity limits of conventional mixers. Current propellant formulations are at the limit of these viscosities. Therefore, more capable mixers or those that do not require the casting process are required to develop new rubbery composite propellant formulations.

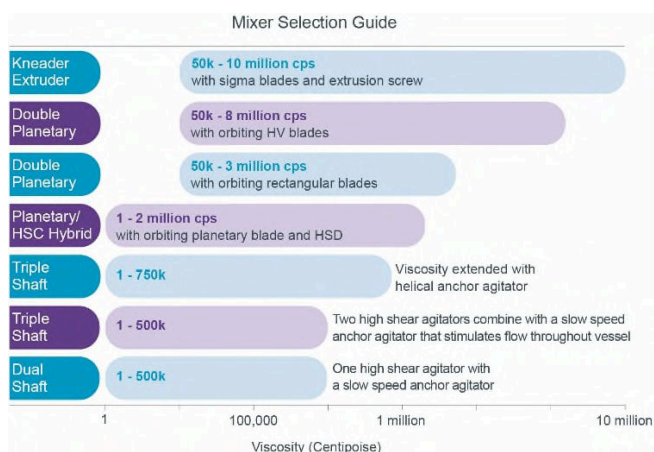


Figure 1. Viscosity limits of conventional mixers. Reproduced from [9]

### 1.1 Resonant Acoustic Mixing

Instead of mixing blades, a Resonant Acoustic® Mixer (RAM) consists of a vertically vibrating spring mounted platform to which a mixing vessel is attached. The oscillations occur at high acceleration (up to 100 G) and low amplitude (up to 14 mm) at the mechanical resonance of the system (approximately 60 Hz)[10].

Resodyn (Table 1) describe the mixing mechanism for composite propellants as “intense material density border interaction through surface disturbances, simultaneous mixing of all ingredients throughout material matrix” [11]. The mechanism of the initially wetting stage of solid components with liquid has been attributed to Faraday instabilities; non-linear waves on the surface of the liquid [12]. On application of high acceleration the manufacturer Resodyn report ‘fingers’ above the surface and ‘cavities’ below it. When subjected to acoustic pressure waves, it is known that air bubbles can influence the flow of surrounding liquid, in a process known as acoustic microstreaming [13], called ‘bubble pumping’ by Resodyn [14–17]. Nance [10][18] computationally validated the presence of these instabilities at the boundary between materials of different densities (i.e. layers of material). However, Nance only modelled the associated vortices and eddy currents for two viscous liquid layers, hydroxyl-terminated polybutadiene (HTPB) resins. The

interaction of particles was not included, and therefore requires further investigation.

Table 1. RAM Processing mechanism reproduced from [11].

Mixing Materials	RAM Processing Mechanism
Powders	Chaotic Collision, Particle Redistribution, Vapor Pocket Movement
Liquids	Intense Material Density Border Interaction through Surface Disturbances, Bulk Mixing
Slurries, Pastes and other Viscous Materials	Intense Material Density Border Interaction through Surface Disturbances, Simultaneous Mixing of All Ingredients throughout Material Matrix
All Materials	Instant and Continuous Bulk Mixing of Materials

Claydon [19] describes the most efficient mixing mode as ‘churning’, whereby the vessel contents couple to the vessel wall, ideally with a ‘no-slip’ condition at the interface [12] [20]. At the start of an oscillation cycle the bulk of the material is given inertia in the upwards direction. When the mixing vessel changes direction down, the bulk of the material does not immediately respond. This creates a velocity gradient between the material adhered to the walls and the bulk material extending perpendicularly across the material towards the center [12] [21]. This results in a bulk rolling motion which provides the shear required for effective mixing [20]. Claydon further discusses that mixing efficiency will rely on the amount of movement in the material and the degree to which the ‘no-slip’ wall condition is fulfilled. By maximizing movement and minimizing wall slip, the velocity gradient (thus shear) will be optimized.

Coguill and Martineau [22] reported that the onset of mixing with increasing acceleration could be described by an empirical relationship related to the vibrational Reynolds number of the system.

Lopez et al. [23] developed a lattice Boltzmann method (LBM) to mimic the RAM, and a discrete element method (DEM) in order to study the impact of particle loading on glycerine/water matrix. Particle image velocimetry (PIV) was used to validate the model. However, peer review of this work was not available for inclusion in this review.

Tanaka [24] reports the optimization of powder mixing conditions by numerical simulation, but the mixing mechanism was not addressed.

Since its development, a large proportion of the research published has tried to understand the mixing but further development is needed before it can be validated and utilised in industry.

Resodyn’s product range has developed since the first laboratory RAMs (LabRAM) was demonstrated in 2006. The products range in both payload capacity and in application. The ‘H’ range are for energetics and meet the standards required for safe processing of sensitive and hazardous materials and range from 1 kg to 420 kg capacity as described in Table 2.

**Table 2.** RAM Models and mixing capacity. Reproduced from [25].

RAM model	Mixing Capacity (kg)
LabRAM I	0.5
LabRAM II	1
OmniRAM	5
RAM 5	36
RAM 55	420

## 1.2 Rocket propellants

Composite solid propellants combine a non-energetic binder (energetic binders are used but are less common) with an oxidiser such as Ammonium Perchlorate (AP). Other ingredients include but are not limited to metal fuel, plasticiser, curing agent, bonding agent and burning rate modifier. The manufacturing is usually by a batch process with multiple stages potentially hazardous for the operator and thus are controlled and monitored remotely. The mixing and casting process are considered the most complex of these stages as it has the greatest impact on the quality and performance of the propellant. The most common method is to mix the material in a batch mixer, such as sigma bladed or planetary, then cast (poured) into moulds. The moulds are then cured [26].

Composites based on HTPB binder are the most widely used. HTPB based composites provide good rheological/mechanical performance, especially at low temperatures, medium cost to manufacture and are well understood, with 50 years of use in the industry [26]. HTPB is a non-energetic polymeric organic liquid which acts as a suspending medium for the solid oxidiser and other ingredients. A polyurethane is formed during the curing process between the hydroxyl terminated polyol and curative.

Curing is required to make the mixture solid with the required physical properties. The most common compositions use the in-organic salt AP as the oxidizer. AP is low cost and has high performance and is used in a wide range of applications. Where increased performance is required, metal fuels are added to the mixture. Aluminium powder (Al) is the common metal fuel used in quantities between 4-17 %w/w, due to its increased performance and cheap cost.

Maximum oxidiser content is desirable to increase the performance of the propellant. Increasing the oxidiser solid loading, reducing particle size and combining multiple particle sizes for better packing to increase the density are all methods increase the oxidiser content and improve performance. Bi-modal and tri-modal packing use two or three different particle sizes respectively to increase the theoretical maximum packing fraction. Tri-modal having a greater theoretical maximum than bi-modal. The optimum theoretical concentration of AP oxidiser would be about 90-93 %w/w, but this cannot be achieved with conventional mixers.

Solid loading (oxidiser, metal and other ingredients) above 90 %w/w increases the viscosity to an extent where the slurry does not flow sufficiently to

mix the ingredients to the required standard [16]. Complex rheological behaviour is reported in solids loading between 86-90 %w/w as the viscosity increases, but the viscosity also changes with shear rate and time [27,28]. This behaviour is apparent in pseudo-plastic fluids and one study reported the maximum solid loading of AP as 85 %w/w in HTPB/AP formulations [29].

Another factor to consider is the mixing time as this is limited by the addition of the curing agent. Once added it creates a curing reaction with the hydroxyl groups of the HTPB, increasing the viscosity with time, limiting the pot life to 4-5 hours before the material is unable to be cast in to the rocket case after this time. The rest of the ingredients are pre-mixed with the curing agent added last to limit the impact.

An increase in shear rate and mixing time are reported to reduce the slurry viscosity, believed to be from shear thinning behaviour of the formulation, caused by the breakdown of large solid particles or of agglomerates and thus reduces viscosity [30]. The breakdown of larger particles making smaller particles, will have more efficient packing as there is now a tri-modal packing arrangement. However, the breakage of particles increases the surface area that needs to be wetted by the suspending liquid, thus leading to an increase in viscosity.

The shear thinning could be due to smoothing of the irregular particle edges reducing jamming between particles [31]. A separate hypothesis is that the shear forces introduce systemization of the particles, and as they become less random and aligned, the viscosity is decreased [32]. Viscosity is also known to decrease with temperature rise, therefore heating the mixture reduces the viscosity and improves flow. Due to the high viscosities, 250,000+ centipoise [33], a high shear mixer such as sigma bladed or planetary is required to achieve a mixture for traditional high solids loading AP/HTPB propellant formulations.

The use of smaller particle sizes and multiple particle sizes will increase the packing fraction and thus the solids loading and viscosity will be increased.

The casting process can be a limiting factor if the viscosity is too great for the mixture to be poured and cast correctly. Mix in-situ is a method to overcome these limitations by removing the casting process. RAM offers an alternative mixing method which may be able to overcome the limitations in conventional mixers for high solids loading and nanoparticles in a highly viscous binder such as HTPB. A higher solid loading will enable better propellant performance, as this should increase the specific impulse and burn rate. Nanoparticles, such as powdered Al can increase the propellant burning rate through an increased packing fraction and can promote smooth burning. These will therefore increase the velocity or range of the weapon system [34].

This paper is a literature review of current published research conducted on RAM and discusses how RAM may effect the rocket propellant industry. 62 journal articles associated with RAM were found. The review will consider the following questions:

- How will the performance and reliability of composite propellants be affected by using RAM over conventional mixers?
- What are the other process/manufacture benefits/drawbacks of using RAM over conventional mixing?
- What will be the impact of RAM on existing qualification testing techniques?
- What is the future for RAM?

## 2 Methodology

This paper is a structured literature review of RAM focussing on rocket propellants, with a particular focus on the high viscosity binder, HTPB, which has been extensively used in composite rocket propellants since 1970 [35]. Initially broader searches were made for RAM, rocket propellants, mixing and HTPB to develop a grounding knowledge and support the writing of the background literature review.

RAM had its first paper released in 2007, unveiling the new mixer [36]. Since then, there has been reasonable research conducted, initially focussed on understanding RAM mechanisms and the effect of equipment parameters on mixing performance. More recently, this has progressed into studies using RAM for mixing but not as the focus of the research. Although RAM is still a relatively new technology this shows how it is already being used for applications in industry, primarily pharmaceutical and energetics. RAM has already gained considerable industrial interest and it is expected this will only grow with an expanding product range to meet specific industry needs and as more research is published. However, there will always be a reluctance from competing industries to publish their findings as it can give them the advantage over competitors. This is reducing the amount of material being published on RAM but this has always been a problem in conducting research.

Throughout this literature review there were an additional 50+ references found in sources which were not accessible. These were either from conferences, which were not published, Resodyn technical interchanges and limited distribution reports. Many of these were used by Andrews [37] who as part of NATO-MSIAC Munitions Safety Information Analysis Centre had access to this information.

Following peer review, additional sources have been included as references to this paper, but as these were conference proceedings and not open literature their findings have not be included in the main body.

## 3 Structured literature review

### 3.1 Performance and reliability of RAM mixtures

The first thing to consider when comparing RAM and conventional mixed materials is the performance and reliability of the finished product. If the mixing produces a different result each time, the resulting material will have differing performance and physical properties and

would not meet qualification testing standards for consistency of results [38]. Therefore, studies comparing RAM and conventional mixing methods will first be considered.

Zebregs et al., compared the density, ballistic properties and homogeneity of RAM and cast-cured rocket propellant of HTPB, ammonium nitrate (AN) and an isocyanate-based curing agent, with an overall solid loading of 81 %w/w [39]. Scanning Electron Microscope (SEM) found no noticeable difference in homogeneity between the two mixing methods. The densities of samples were found to be within 1 % using Helium gas pycnometry. A chimney burner determined that the RAM mixed propellant was within 5 % of the burn rate (2-10 MPa pressure) for the Baker Perkins mixed propellant. There were no burn rate catalysts used in the experiment. Overall, there were no discernible differences between the two products.

Nelson & Cross, [40] conducted a comparative study on a 125 g hydroxyl-terminated caprolactone ether (HTCE) binder, AP and Al mixture made by LabRAM and Baker-Perkins (BP) mixers. The viscosity, burning rate and tensile properties of the final products were compared. Burning rate and tensile strength were comparable between methods. End viscosity was significantly greater for RAM (21.5 kPs @35.5 °C), than BP (7.5 kPs @49.3 °C). The reason for the lower viscosity for the BP produced material was not stated but may be due to increased processing temperature. The greater viscosity of the RAM mixture could impact its ability to cast into the casing. They proposed mixing in-situ to overcome this limitation. The mixing time was noted to be shorter for RAM, 30 min compared to 100 min for BP. No agglomerations were seen in the RAM mixture. Given that this was one of the first published papers on RAM, it is possible/likely that the mix process and time for the RAM was not optimised and could have been considerably quicker. Especially as the mix time for the BP mixer seems quite short suggesting that the composition should be easy to formulate.

Vandenberg & Willie, [41] conducted a comparative study of ultra-high performance concrete (UHPC) mixed with table-top paddle mixer (TTPM) and LabRAM. The microparticles, high volume content of filler (30-50 %w/w), highly viscous mixture and desire for homogeneous mix for better performance make this a comparable study to rocket propellants. The RAM product showed increased rheological properties (viscosity) but improved mechanical properties over TTPM. RAM mixed UHPC had a 30 % increased compression strength after 3-day and 20 % after 28-day observations. After 56 days the RAM product had a compression strength on average approx. 200 MPa compared to 165 MPa for the TTPM prepared mixture. This was attributed to a more uniform mixing energy in RAM which enhances hydration and reduces air voids. The use of vacuum could have improved the mixing quality and performance further, as it could increase the mix homogeneity.

Rumeau et al., [42] found an improved mixing quality of epoxy resin and titanium oxide powder with



LabRAM compared to an unspecified mechanical mixer. The mechanical, topographical and thermal properties of the RAM mixture were found to be within specification. The average relative standard deviation (RSD) of the ultimate tensile strength was found to reduce from 18.6 % (conventional mixer) to 5.1 % (RAM). The reduction in RSD shows good reliability and reproducibility of RAM as a mixing technique and that it produces a higher quality product. Although it is difficult to validate the experiment due to the lack of detail on the exact mixture and mechanical mixer used.

Park et al., [43] used LabRAM to successfully fabricate NiO-yttria stabilised zirconia anode supports for solid oxide fuel cells. Conventional ball-milling (BM), addition of plasticiser and binder and de-airing the slurry would be a >72 hour process, whereas it took about 30 min with RAM (x144 faster). Quantification of the product was conducted using 3D reconstruction technique and electrochemical performance was tested, both finding that the mix was highly homogeneous and statistically identical to BM. The product also showed good long-term stability of over 300 hours, making RAM a desirable alternative to BM in this field.

The final two studies consider mixing of liquids with nanoparticles in LabRAM. Leung et al., [44] compared RAM and milling for nanoparticles of Naproxen, an active pharmaceutical ingredient (API), and a polymer. Different stabilisers were tested and average particle sizes of the nanoparticles were compared between the two techniques. For stabiliser, PVP K28-32/SDS, the D90 average particle size was found to be 276 nm and 2510 nm respectively for RAM and milling. Significant aggregation was also observed in the milling product under optical microscopy. The RAM produced formulations were stated to have better physical stability, although values were not stated to support this. The better stability was due to lower shear, no contamination from blades or container, uniform energy and mixing and less temperature increase to the RAM mixture. This amounts to a more stable product with better physical properties. In addition, RAM was able to mix higher viscosities, with concentrations up to 50 % (500 mg/ml) naproxen drug loading, whereas milling was limited to 20 % (200 mg/ml).

Nellums et al., [45] reports on a comparative study of the nanothermite aluminium-bismuth(III) oxide (Al/Bi<sub>2</sub>O<sub>3</sub>) mixed by ultrasonics and LabRAM. Ultrasonic mixing was limited to a very low solids mixing of 0.6 %v/v and RAM up to 50 vol%. Comparison and consistency of electrostatic discharge (ESD) times were used to determine the quality of the products. The ignition delay, for ten experiments, for thermite ultrasonicated in hexanes was 106±7 µs, compared to 95±3 µs for thermite with N,N-dimethylformamide (DMF) at 40 %v/v using RAM. The 30-50 %v/v solid loaded RAM mixtures had the most consistent ignition delays, and smallest aggregates (2.3 µm nom. aggregate size), overall producing an improved mixing quality. In both these cases RAM was able to produce mixture that were beyond the capability of the conventional mixer.

In each of the seven comparative studies reviewed RAM has either performed as well as the conventional technique or better. A more homogeneous mix producing a better quality product was seen in four of the studies; Vandenberg & Willie, Rumeau et al., Leung et al. and Nellums et al. Other reported benefits of RAM were, faster mixing time, greater homogeneity, fewer agglomerates, less contamination from blades/impellers, lower shear and a more consistent mix. RAM was able to mix higher quantity of nano particles in two of the studies Leung et al. and Nellums et al. and higher solids loading in Vandenberg & Willie and Nellums et al. A greater viscosity of the RAM product was reported by Nelson & Cross, which could be problematic for the casting process. This observation was not seen in any other study.

The next subsection considers the sources which have conducted testing on materials which were not possible with conventional mixing methods, such as higher solids loading or nanoparticles which make the material too viscous for high shear mixing. Two such studies have been conducted at Cranfield University. Firstly by Brodier, [34] who evaluated the addition of aluminium oxide nanoparticles into a representative HTPB based propellant mixture on LabRAM. Sugar was used as an inert substitute for AP and the nanoparticles were added to samples of 3, 5, 7, 9 (%w/w) and a baseline sample with 0 % nanoparticles while keeping the solids loadings consistent at 60 %w/w. An increase in viscosity was found in the 7 %w/w and 9 %w/w samples, which is to be expected from the addition of nanoparticles. Some limitations observed were the poor coating or particles and presence of agglomerations, which differs to the general reporting of RAM. In this experiment the most intensive mixing was set at 70 g for 7 min which may not have been long enough or intense enough to break down the agglomerates as other studies have used high acceleration, longer mix times or pre-wetting to achieve this. Also, the ability of RAM to coat particles has been well reported, indicating the observations in this paper may be due to the specific mixing times and intensities not being optimised.

The second study was conducted by McCloy, [20] who investigated the effects of solids loading on viscosity using LabRAM. The experiments were carried out on HTPB, dioctyl sebacate (DOS) plasticiser and coarse and fine sugar as substitutes for AP. The material viscosity peaked at the maximum theoretical packing fraction at solids loading of 60-70 %w/w. With solids loading above 70 %w/w the viscosity dropped and it was observed that the sugar formed agglomerates as it was not properly wetted by the HTPB. The paper highlights how sugars are not perfectly representative of AP as the material characteristics and flowability will be different, the crystal shape of the sugar particles is more angular and irregular compared to more rounded AP.

As previously reported, AP has been mixed with HTPB to greater solids loading levels (>80 %w/w) with conventional mixers and has a theoretical maximum solids loading at 90-93 %w/w [26]. Increasing the temperature during the pre-wetting phase has been

reported to reduce binder viscosity and better able to wet the solid particles [46]. Unfortunately, this study did not discover if RAM was able to mix higher solids loading than conventional mixers and further research is required in this area.

From the studies considered in this section, RAM is able to coat or mix higher concentrations of nanoparticles with liquid binders than conventional techniques in Leung et al., Nellums et al. and Brodier. While this research area is still in its infancy there are promising results and with optimisation RAM is likely to develop even greater nanoparticle formulations. The ability of RAM to mix greater solids loading is less clear, with only McCloy investigating this directly and reporting challenges with mixing at higher solids loading. Further research in this area is needed to access the solids loading limitations of RAM for HTPB based propellant mixtures. In addition, experiments will need to involve energetics mixture with the oxidiser.

While none of these studies specifically consider mixing of HTPB/AP/Al, the ability of RAM to produce reliable and highly homogeneous mixes make RAM an attractive option for research or manufacture. RAM has mixed nanoparticles and higher solids loading than conventional techniques indicating that RAM will likely be able to produce previously un-mixable HTPB based propellants.

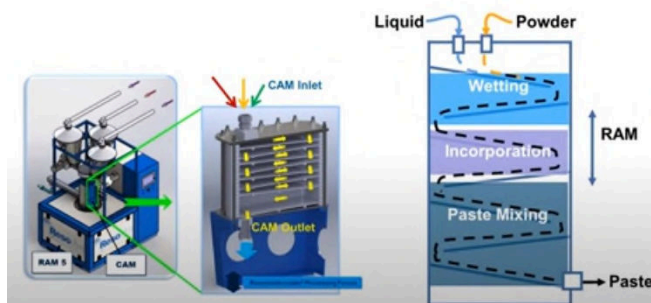
Looking beyond the published literature this last section will consider the most recent RAM development by the US DoD and have presented their results in a series of webinars. As previously discussed, mix in-situ on RAM and CAM are two areas. Unfortunately, there were no associated reports with further detail, but these studies have been conducted in conjunction with Resodyn and are considered some of the leading work in the field.

First is a comparative study on the production of the granular pyrotechnic, Magnesium/Teflon/Viton (MTV), through RAM and conventional methods [47]. The key findings from using RAM will each be discussed. A 95 %w/w was pourable as granules immediately after mixing, making this a low waste process. Process has been scaled from 25 g to 100 g scales with near identical results. Cost comparison was made between the two processes and found in terms of materials and labour RAM showed a 35 % saving per batch and completed this in 5 h compared to 10 h on conventional. Overall RAM was safer, produced less waste, had less environmental impact, was 35 % cheaper per batch and halved the time of production.

The second study produced polymer bonded explosives (PBX) shaped charge warheads mix and cast with a bladed mixer (17 made) to mix in-situ with RAM (18 made) [48]. The specific PBX was not stated. Composition analysis determined that the global standard deviation of the solids content was  $\pm 0.217$  %w/w and  $\pm 0.09$  %w/w for the two formulations tested. The military specification required is  $\pm 2.0$  %w/w therefore this was very good mixing.

Mix in-situ took 0.77 man hours per warhead compared to 1.53 for mix and cast. Mix in-situ required 79 % less solvent and produced 63 % less hazardous

waste, making it more cost effective and environmentally friendly. One of the other benefits of mix in-situ is that the buildings explosive licence only needs to be for the amount being mixed at that time which is smaller each run with RAM compared to larger batch mixers. For this reason, the Quantity Distance (QD) will be less and have less of a safety impact on other personnel and buildings.



**Figure 2.** Schematic of continuous acoustic mixer. Image taken from [48].

Continuous acoustic mixing (CAM) and Clean in Place (CIP) are the focus of the last study and shows the biggest step forward in recent RAM development. Figure 2 shows the CAM-CIP configuration on the RAM 5 (36 kg capacity for batch production) and how the mixing process is conducted with the viscous ingredients moving through the CAM by gravity [48]. This configuration is able to process viscosities  $>1,000,000$  cp at room temperature at a rate of 3.0 kg/min. The level of acceleration is increased to mix higher solids loaded or higher viscosities and these need to be optimised for each mixture. The PBX mixed has the solids loading percentage tested by thermal gravimetric analysis and was well within the required  $\pm 3$  standard deviations.

To CIP, the CAM system is run with just water and air. The agitation of the water causes aggressive cleaning with only 5 kg of material waste and 9 litres of aqueous waste from however long the mixing was conducted for. The amount of waste is therefore not linked to amount of material produced, where it would be in batch mixing. This also removes need for organic solvents and the operator is not exposed to hazardous solutions.

### 3.2 Manufacturing benefits and drawbacks of using RAM over conventional mixers

While an improved performance and reliability of a product are highly desirable for a new manufacturing technique there will be other benefits/drawbacks which need to be considered to understand if the technique should be implemented. Safety and environmental factors will be of high interest but the leading factor will be cost. The benefits and drawbacks will be considered for three main areas; (i) research and development, (ii) ease to scale up and (iii) large-scale production. The benefits and drawbacks applicable to all three of these areas they will be discussed first before going into the individual sections.

There are four consistently reported benefits of RAM that impact all phases of a product development. These are (i) shorter mixing time, (ii) more homogeneous mix, (iii) gentler or low shear mixing and (iv) less waste. Similar or improved product has been widely reported but has already been addressed in the previous section of this review. Each of these will impact the manufacturing process and therefore cost of the final product. Reduced mixing time is one of the main attractions of RAM and has been reported in ten of the studies [24,37,39,40,42–44,49–51]. It should be noted that Andrews [37] produced a literature review rather than experimental study, therefore the reduced mixing time was reported from other studies.

Park [43] reported the mixing time of anode supports for solid oxide fuel cells was reduced by x140 to 30 min with RAM compared to 70 hours for ball-milling. Similarly, Batmaz [51], reported an >80 % reduced in time to pulp and sterilise banana puree in the food industry. Rumeau [42], saw a reduction of mixing time from 15 h to 6 min when comparing powder mixtures between LabRAM and conventional mixers (double cone blender in this example). There have not been any reported occurrences where the mixing time has increased using RAM. However, Sharma [52], identified that overmixing of dry particles beyond the optimal time caused heat accumulation and particle attrition. This highlights the importance of optimising the process as the particle attrition will change the particle sizes, which will then affect the performance and mechanical properties.

A more homogeneous mix can improve the performance of a mixture, but it will also make the mixture more consistent between batches. Therefore, the performance and properties of that mixture should also be more consistent, which was reported in five of the studies [24,43,44,49,50]. Osorio & Muzzio [50], found that mixing of low concentrations APIs could be achieved in a little as 30 s and an increase in acceleration or time did not improve mixing performance. Park [43], reported solid oxide fuel cells that use a liquid binder and therefore similar type of mixing to HTPB based propellants. In this study the mix was high homogenous and statistically identical microstructures. None of the studies highlighted a reduction of homogeneity with RAM compared to conventional techniques. Numerous studies did identify agglomerates in the final product, and this will be discussed in more detail later in this review.

The gentle mixing mechanism of RAM is a common theme in the literature. While some of the studies state this specifically, it is implied in many more studies [39,44,49,50,53]. RAM is described as low shear in these studies, and this has its positives and drawbacks. From a positive perspective this can limit the impact of the mixing on the particles and therefore not change the particle sizes which is evident from conventional high-shear mixers. Zebregs [39], observed through SEM that the HTPB based propellant particles were not fractured or damaged during the mixing process in RAM. Leung [44], reported that the lower shear of RAM over traditional milling reduced the risk of

agglomerations forming as milling causes physical stability issues.

The energy transfer from RAM is uniform across the entire contents of the mixing vessel compared to a grinding chamber in a mill than has areas of at least 6.5 times higher intensity. The maximum intensity of mixing imparted in RAM is therefore lower and this is proposed to be why the nanoparticles experience less stress and have improved stability. Hope et al. [53], reported that with higher mixing intensities (50 g and 100 g) sucrose formed an increased amount of agglomerates however the crystals did not experience major shearing or fracturing. This lack of shearing potentially makes RAM safer and thus more attractive for more sensitive energetic formulations, especially at lower mixing intensities [54,55]. Safe mixing of sensitive explosives using LabRAM has been demonstrated for nanothermites [45].

The drawback is that the lower shear does not breakdown the agglomerates within the mixture. This can be managed in different ways including pre-mixing, pre-wetting or intensity of mixing which is related to application of vacuum in RAM. The terms pre-wetting and pre-mixing both have the same intent of making a semi-mixed state prior to intensive mixing and the terms appear to be used interchangeably. Pre-wetting has been seen more often in these studies as it refers to the liquid plasticiser specifically, in making sure the liquid is in contact with a single solid or all of them. The use of the two terms will be described separately for clarity.

A pre-wetting stage at lower accelerations have been used to take the initial ingredients to a partially mixed condition [39,46]. Andrews [37], highlights the importance of the pre-wetting stage and how it impacts the forming of agglomerates and the mix homogeneity. The intent of pre-wetting is similar to pre-mixing in that it evenly distributes the liquids across the different solids, which is of greater importance if there are different levels of solubility of the solids [56].

Yew [57], determined that pre-mixing of the binder and plasticiser of a HTPB, DOS, coarse sugar and fine sugar mixture could reduce mixing time from 60 min to 45 min to reach the same standard deviation of 0.27 AU/Mass. But after only 5 min a standard deviation of 1.0 AU/Mass was reached with pre-mixing. In this experiment the binder and plasticiser were pre-mixed by hand for 5 min before the addition of the other ingredients and mixing on RAM. Pre-mixing is also common in conventional mixing and has significant impact of the final product produced.

In Nelson & Cross [40], a formulation of HTCE binder was mixed with AP and Al on LabRAM. Mix B, with a particle surface area of 1.65 m<sup>2</sup>/g, was mixed at a stated 100 % intensity for 20 min intervals. The acceleration was stated as 40-50 g throughout the experiment but did not state the specific amount applied at each stage of the process. Instead the intensity was stated, which will change depending on amount of material in the vessel. Vacuum was not applied throughout all experiments. Half the total solids were mixed for 20 min then the remaining half of the solid

ingredients were added and mixed for a further 20 min. At the 20 min and 40 min points agglomerates were observed. After a total of 60 min of mixing at 100 % intensity the agglomerates were no longer observed. The lack of agglomerates was confirmed through optical microscopy. Another test was conducted in a smaller mixing vessel, with mixing intensity set at 50 % for 60 min. No agglomerates were observed in this experiment. These examples show how the intensity of the mixing and the mixing time can contribute to breaking up of agglomerates, also known as de-agglomeration. The lack of applied vacuum throughout the experiments may have also been a factor in the original formulation of the agglomerates or the length of time required to break up the agglomerates.

McCloy [20], demonstrated that the application of vacuum reduces the required acceleration/intensity required to initiate a churn in the mixing. For an 80 %w/w solid loaded HTPB, DOS, coarse and fine sugar mixture it was shown to churn at 50 g acceleration (50 % intensity) when vacuum was applied but no churn occurred without the vacuum at 50 g. The intensity required to churn without vacuum was not tested. The effect of vacuum was explained from the air bubbles compressing and expanding when subject to acoustic mixing due their relatively low shear modulus. The distorted bubbles cause a flow around the surface of the bubble and aid in mixing.

Yew [57], showed that the mixing time of a HTPB, DOS, coarse sugar and fine sugar formulation can be reduced from 60 min to 2.5 min with the application of vacuum after 1 min of mixing on LabRAM. A standard deviation of 0.41 AU/Mass was reached after 2.5 min. Therefore, the lack of applying vacuum may account for the formulation and longer time to mix and breakdown of the agglomerates in the experiments conducted by Nelson & Cross [40].

The last of the benefits across all areas of product development is the reduced waste produced by RAM compared to conventional mixing techniques. This will have the greatest benefit in large scale manufacturing, but also assists in smaller batches. When a highly viscous formulation is mixed in a bladed mixer there will be a considerable amount of residue on the sides of the container and on the mixing blades [44]. Therefore, more material is lost in the conventional process and further to this more cleaning solvents will be needed to clean the vessel for future use [39,46]. The clean-up phase will have costs in cleaning materials, protective equipment, operational down time and cleaners time as well as the associated health risks [58]. The environmental impact of the explosive residue and solvents will also be reduced which is an important consideration in modern industry.

RAM also offers the ability to mix in-situ. Where the casing is attached onto the RAM with the ingredients poured in and the RAM will then mix the formulation in the case. Jubb [46] successfully made and fired rocket motors which were mixed in case. Unfortunately, there was limited information on the formulation ingredients, process and performance reported. This has the benefit of effectively zero waste and clean-up costs. Dependent

on space and mass, multiple casings could be mixed simultaneously, which allows a greater throughput and subsequent cost saving. Mix in-situ or in case will be discussed further later in this review.

Each of the development stages; Research and Development (R&D), scaling and large-scale production will now each be considered. Followed by how safety is considered across each of these stages.

R&D is an important stage for pharmaceuticals and energetic formulations. The introduction of RAM has unlocked an ability to greatly increase the number of different formulations to be initially tested and developed. There are two main benefits of RAM in R&D.

Firstly, different size vessels can be used to optimise mixing depending on the amount of material required to be mixed. This is not the case with conventional mixers [41,50,52]. Osorio [50], did identify that the fill level did affect the temperature of the final product. Mixing time was also increased with a greater fill level, which is to be expected as there is more material to be mixed. As the fill level does not affect performance this means that different volumes of material can be prepared which has multiple benefits. This allows smaller samples to be prepared, which may be desirable for safety (energetic formulations) or to save on material costs. Leung et al., [44], was able to prepare nanosuspensions of 1-2 mg each in a 96-well plate. This allowed both an ideal sample size and a higher throughput of samples. Am Ende [56], similarly suggests that multiple vials of co-crystals can be prepared simultaneously, but this was not conducted in their experiments.

The second benefit for R&D is that RAM is relatively simple to use and the mixtures can be reliably reproduced. The RAM and associated ancillaries are relatively simple to use and do not require specific training by Resodyn to use or need considerable experience to use effectively. However, optimisation of the mixing process is more difficult as it requires understanding of the materials and correct setting of the accelerations for different periods. This may take many attempts to optimise. The software that comes with RAM can record the acceleration, vacuum and other parameters and then this can be used as a template to make future mixtures. These templates can also be sent to manufactures or other laboratories who will be able to reproduce the material. This could be for larger scale production or for further testing in another location/country, aiding international collaboration. This assumes that if the same parameters are used on RAM then the same product is produced.

While this may be true for the same RAM, Claydon et al. [19] reported there have been less consistent results from running the same process on different LabRAMs due to differences in tolerance between each mixer. As the RAM products have developed over the last 10 years and into the future, the reliability and consistency between RAM units is expected to improve considerably.

The next step of production is the scaling process. This is usually conducted in stages from grams to 100s of grams before kg and large-scale production.



As discussed, the ability to record the system parameters and then repeat the mixture makes the scaling process relatively easy. This is applicable to larger batches in the same size RAM or scaling up on a larger RAM. Resodyn has stated that RAM is amenable to scale-up as the mixing mechanism is consistent between different scales and energy scales up with the increased capacity in the larger RAMs [36,59]. Am Ende [56], produced co-crystals in batches of 150 mg, 1.5 g and 22 g without needing to adjust mixing time. On the 1.5 g and 22 g batches there was 0.2 % and 0.4 % or the surface area respectively which had not crystallised. Future work is planned to scale up sample sizes to 100 g and 10 kg to determine if there are any differences in process or performance.

Nagapudi [60], also scaled up co-crystals from 400 mg to an 80 g batch. The 80 g batch had a net yield of 94 % which was achieved after 2 hours mixing which was the same as the smaller 400 mg batch. Both co-crystal studies show how co-crystals can be scaled up without changing the mixing times or performance on the same LabRAM apparatus. While this is not directly comparable with high viscosity mixtures it shows the scalability of RAM. Jubb [46] has used a LabRAM to make 1 g, 5 g and 100 g of composite propellants. Then later scaling up the process to make 180 g propellant mixture in a single batch. In 2014, making a 5-inch rocket from 14 batches of propellant made on a LabRAM, which was successfully fired. In 2020, Jubb et al. [61] reported the manufacture and firing of a 7-inch 49 kg composite propellant motor was using 7 batch mixes on an Omni-RAM. Their work shows how effective RAM is for different sized mixtures. Unfortunately, there have not been any other studies which have looked at the scaling up of mixtures between RAMs of different sizes. Jubb [46] has reported to be formulating propellant with a LabRAM IIH (1 kg), Omni-RAM (5 kg) and soon a RAM 5 (36 kg), with a view to publishing comparative work in the near future.

Larger scale production is where the cost factors are most apparent. There are significant costs in the purchase and set up of a larger RAM, however, these are offset by the numerous benefits; shorter mixing times, less cleaning, less waste material and solvents, less environmental impact and ability to mix new formulations have all be discussed. A RAM can also be used for multiple explosive mixtures including high explosives and propellants and making it a versatile instrument. While there are examples of large scale use, there are no examples in the journal articles of large-scale production or the use of a larger RAM on the 5 kg to 420 kg scale. Even so, there are factors such as temperature increase and safety which will need to be considered at this scale. These will need to be tested during larger scale testing to ensure there are not safety or performance impacts.

Now each of the production stages have been discussed the safety aspects will be considered, which is always a concern with energetic mixtures. The main areas to consider with RAM is the risk of ESD, safety of operator during operation and safety of cleaners. When mixing powders, especially nanoscale particles, there is

a risk of ESD causing an ignition [45,62]. This was an initial concern with using RAM and Andrews [37], reported that although ESD was detected, the charge could be dissipated using earth bonding onto the mixing vessel (if it is conductive). Stainless steel and titanium vessels were both used for this but being opaque meant that the mixing process could not be observed. Andrews also reports on another study, which was presented by Beckel at the 6th Resodyn Technical Interchange, that the ESD risk from RAM is comparable to that of conventional mixers. Further information would be desirable to understand what experiments were conducted to determine this assessment.

For the safety of the operators when mixing, the desire is to have remote monitoring and control, therefore the operator can be at a safe distance with suitable structural protection. To achieve this, there need to be sensors and monitoring systems in place, which is normal for energetics mixing but may not be in place for all sites using RAM [37]. Many sites have incorporated temperature monitoring or control. Monitoring through thermocouples incorporated into bespoke mixing vessels or by using heating/cooling sleeves which have the additional benefit of being able to control the mixtures temperature. Larger versions of these have been developed and incorporated into the production scale RAM products. With the larger vessels it is more difficult to control the temperature from the outside and this can cause a safety issue. Multiple thermocouples can be inserted through the lid to measure temperature throughout the formulation and helps mitigate this risk of overheating. The Falcon Project UK have incorporated a hardware E-stop to their RAMs to remotely remove power to the unit if required [46]. This is now standard for the hazardous approved RAM products.

There are two main risks in cleaning mixing vessels. Firstly, from the risk of an explosive event during cleaning and secondly from the health risk from the mixture and hazardous solvents used for cleaning. As previously discussed, mix in-situ removes both these risks but in a batch process cleaning will need to be carried out each time. With RAM, the lack of internal moving parts and blades reduces the material to be cleaned, amount of solvents required and time of clean, making RAM a potentially safer process than conventional mixing. Clean in place (CIP) conducted remotely also offers a safer option and this has been demonstrated with the continuous acoustic mixer, which will be discussed in more detail later in this review.

### **3.3 How will the qualification testing of explosives be affected by the introduction of RAM**

This section will be considering how RAM offers an opportunity to redefine the process for qualification testing of RAM produced formulations. There have not been any published studies directly looking at this question yet, but it is a known problem in the field [37,46]. For this reason, this section will use wider sources and those from other industries who are currently considering this same problem.

Allied Ordnance Publication 7 [63], provides the core tests that NATO nations require to qualify an energetic material for a specific role. These tests are thought to be broadly sufficient for existing propellant formulations manufactured on a RAM. Andrews et al. [64] discuss that for propellants that can only be produced on the RAM (e.g. very high viscosity cast-cure formulations) there may be a requirement to carry out testing that exceeds that of current capability of test instrumentation. If the batch size on the RAM was large enough then there would likely be no need to change the process for batch to batch certification and qualification. Andrews goes on to discuss that mixed in case processing will be a more difficult process to consider. As now it is difficult to differentiate from traditional energetic material qualification and type qualification in the intended role; due to fact that your energetic material is being filled directly into its end use item. To consider the batch size of one as being suitable for service, then assurance must be gained through process control as well as ingredient control.

Currently, defence industry relies upon destructive all up round trials testing that often provides inconclusive datasets. This does not provide any real confidence about the process. Furthermore, testing often occurs late in the product development lifecycle, where the impact of any emergent risks has the potential to be hugely costly.

The qualification of new processes or materials is often cited as a barrier to bringing new product technologies into service. In order to reduce timelines and costs the UK defence community is actively pursuing novel means of introducing innovation into product offerings.

The UK defence community has started considering a new framework of repeatable and reliable testing to form an understanding of the energetic material [65]. It has identified the key to this was to define and gather critical material information. With priority on combustion/burn characteristics, material definition (chemical and physical properties) and bare and confined performance output. This shifts from the purely output focussed testing to a greater understanding of the base materials to then inform their output performance.

This reinvigorated approach to qualification is driven by a need to develop a better understanding of the system behaviour rather than relying upon system trials results. By investing in small scale material tests the community is able to validated predictive models that can be substituted for the current attribute driven trials activities. This move to a more scientific approach to qualification has been branded smart or agile qualification. The general work packages included in the ongoing UKs 'agile qualification' activities will focus upon the characterisation of energetic materials around:

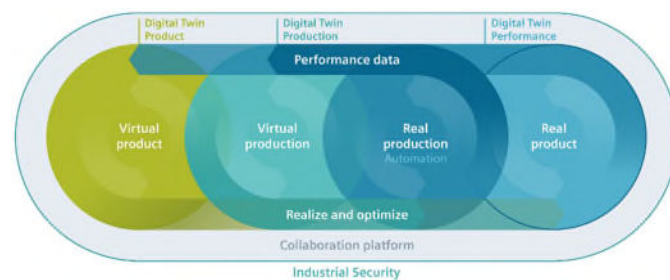
- key performance, safety and life parameters
- provision of reliable and repeatable test methods
- provision of validated productive modelling
- an agile approach to qualification definition (allowing best in class testing to be adopted)

- the introduction of pan enterprise assurance models to ensure the appropriate level of assurance for weapons systems and
- the derivation of digital twin and digital thread frameworks to enable 'overnight' digital design and qualification activities with improved confidence over design margins.

With this understanding comes the potential to improved service life predictions for advanced formulations – potentially reducing the number of weapon systems prematurely disposed of due to a lack of understanding of the residual service life.

RAM provides a process controlled reliable and repeatable manufacturing process therefore, supporting agile qualification testing and potentially negating the need for batch to batch certification or testing every munition for mix in case. By conducting small scale testing of the material energetic properties during the development phase, the output performance and safety implications can be predicted for the larger scale material. Although not yet validated, scaling is likely to not affect the material properties due to the nature of RAM mixing.

There is an intent for change in the energetics industry, but how is this implemented? This is best addressed by considering the lessons learned from other industries. The world is venturing into Industry 4.0 and similarly many industries are moving towards Formulations 4.0. They use the data gathered to generate a digital twin. An example of a digital twin is shown in Figure 3 and shows how the digital twin can be applied to energetics with the base ingredient properties (product), manufacturing and mixing (production) and ballistic and mechanical properties (performance) all being recorded and modelled. To achieve this level of modelling representation for the entire process it will require many models all incorporated together.



**Figure 3.** Digital twin example from Siemens Global. copyright ©Siemens AG. [66]

There are different types of model that can be implemented which range from no, limited or full linkage into the manufacturing equipment. Due to the recording systems on RAM it would be relatively simple to link this into live monitoring model. However, RAM modelling is not mature (section 1.1) and not at a stage where this could predict performance outputs from different mixing parameters.

The pharmaceutical industry has faced these challenges in the last ten years and evolved the use of complex models into the process development. Rogers

[67], produced a paper on the challenges and opportunities this type of modelling enables to their industry. Specifically looking at how mixing models enable the ability to conduct dynamic simulations to assist in process design and optimization. The study concluded that further work is required in the flow behaviour in micro-scale mixing to improve the predictive ability of the model to determine blend homogeneity.

Singh [68], describes how integrating process analytical technology (PAT) into the manufacturing enables automatic control of end product quality. Meeting the quality by design paradigm required by the Federal Drug Administration (FDA). They used Near Infra-red spectroscopy to determine powder bulk density and used PAT for feed-forward and feedback control to produce a pre-defined end product of the required quality. This system was planned to be incorporated into a pilot-plant for further testing at a larger scale. These types of monitoring systems are in place which are reducing R&D times to reach drug certification, dramatically improving efficiency and reducing costs. Certara® [69], who specialise in model-informed drug development, state they have supported through their software or services 90 % of the new drug approvals in recent years.

The next challenge to consider is how to monitor RAM during mixing. As discussed, the temperature and mixing parameters can be monitored but this is likely not enough to produce the data required for certification. If the modelling was mature enough to only require mixing parameters to validate the mixture then this would not be required, but until that point it would be desirable to have monitoring systems in place. Optical microscopy, SEM and near-infrared spectroscopy have each been used for determining performance of RAM mixtures [39,40,50]. But each of these are not practical during mixing as sampling from the mixture is required.

Ultrasonic transmitters have been used on the piping following mixing on the CAM to determine if the correct level of mixing has been achieved [48]. X-rays can be used post- production for cast or mix in-situ to identify any cracks in the propellant or bonding issues between the propellant and the casing, but could also be used during mixing. Michalchuk [70], used synchrotron X-ray powder diffraction (XRPD) to conduct real-time monitoring of co-crystal generation. Similarly, Halls [71], used X-ray attenuation for non-destructive homogeneity testing in liquids, which could be suitable for in-situ monitoring of RAM. While each of these X-ray methods have not been considered before in the context of in-situ monitoring of a RAM mixing vessel, they show areas of potential future research as an aid to modelling and understanding of the mixing mechanisms.

## **4 Conclusion**

RAM is able to mix the current level of solids loaded composite propellants with numerous benefits including shorter mixing time. RAM and CAM can mix higher

viscosities than conventional mixers so will allow higher solids loading, including formulations with nanoparticles. The solid propellant Industry could therefore shift focus to new oxidisers with a higher packing fraction with improved performance. This could facilitate the new generation of RAM produced composite propellants which have improved performance and mechanical properties.

With RAM, new propellant formulations will be able to be tested at R&D level and scaled up more quickly than conventional methods, so the rate of new formulations being developed is expected to increase significantly, opening opportunities for different oxidisers, binders or other ingredients.

The reduction in production times and overall cost should increase the pace of research in this field. As RAM becomes more common in R&D institutions and in wider industries, it will also increase the rate of product development. While CAM-CIP and mix in-situ have been conducted in some studies, these are expected to be adopted more widely in the future. With this comes the monitoring in-situ which is also expected to increase as the energetics community moves towards more flexible and modelling based production. Where RAM offers a real opportunity to establish a new energetics qualification test criterion as it is a new technique which benefits from its ability to record and repeat the mixing process. Further developments in modelling and monitoring in-situ are required to enable this. X-ray and ultrasonic based monitoring methods have been suggested.

As RAM is relatively immature in its use in the materials industry there is still a considerable amount of research required for the benefits to be widely utilised. Key areas for future R&D are:

- Understanding the mixing mechanisms with different system parameters and how this impacts output performance
- Development of models to aid understanding of the mixing mechanisms
- Comparative studies on small and medium/large scale production using RAM focusing on how scale up impacts material characteristics and output performance
- Methods for in-situ monitoring and linking this to output performance
- Tools for predicting output performance from different ingredient and systems parameters such as using digital twin concepts. Building towards validated productive modelling to enable agile qualification of energetic materials
- Validation of mix in-situ techniques and how this affects qualification testing
- Further novel uses or modifications of RAM such as CAM or mix-in-situ to widen its different applications
- Continued sharing of information between Resodyn, R&D and industry to build the community knowledge of RAM

Overall RAM offers numerous benefits to mixing existing and new materials with large savings in time and cost and being safer and more environmentally

friendly. Composite propellants have been the focus of this study, but RAM is expected to increase in use in both the energetics community and wider mixing industries.

## Acknowledgements

Chris Wright would like to thank the Ministry of Defence for funding the Explosives Ordnance Engineering MSc enabled the production of this literature review. We would like to thank Clare Pratchett and her team at Cranfield University-Learning Services for the re-production of the diagrams.

## Symbols and Abbreviations

Al	Aluminium powder
Al/Bi <sub>2</sub> O <sub>3</sub>	Aluminium-bismuth(III) oxide
AN	Ammonium nitrate
AP	Ammonium perchlorate
API	Active Pharmaceutical Ingredient
BP	Baker Perkins mixer
BM	Ball milling
CAM	Continuous Acoustic Mixer
CIP	Clean in place
DMF	N,N-dimethylformamide
DOS	Diocetyl Sebacate
ESD	Electrostatic discharge
FDA	Federal Drug Association
GDP	Gross Domestic Product
HTCE	Hydroxyl-terminated caprolactone ether
HTPB	Hydroxyl-terminated poly butadiene
LabRAM	Laboratory Resonant Acoustic Mixer(s)
LESLIE3d	Large Eddy Simulation with Linear Eddy modelling in 3 dimensions
MTV	Magnesium/Teflon/Viton
MSIAC	Munitions Safety Information Analysis Centre
PAT	Process analytical technology
PBX	Polymer Bonded Explosives
QD	Quantity Distance
R&D	Research and Development
RAM	Resonant Acoustic Mixer(s)
RSD	Relative standard deviation
SEM	Scanning Electron Microscope
TTPM	Table top paddle mixer
UHPC	Ultra-high performance concrete
XRPD	X-ray powder diffraction

## References

- [1] World bank organisation, "Manufacturing, value added (% of GDP)," **2020**.
- [2] The Manufacturer, "UK Manufacturing Statistics," **2020**.
- [3] H. Kagermann, R. Anderl, J. Gausemeier, G. Schuh, W. Wahlster, *Industrie 4.0 in a Global Context - Strategies for Cooperating with International Partners (ISBN 0773528490)*, **2016**.
- [4] E. L. Paul, V. A. Atiemo-obeng, S. M. Kresta, *Advances in Industrial Mixing - A Companion to the Handbook of Industrial Mixing (ISBN 0-471-26919-0)*, John Wiley & Sons, Inc., Hoboken, New Jersey, **2016**.
- [5] F. S. G. Abadi, The Role of Rheology in the Flow and Mixing of Complex Fluids. Master of Philosophy Thesis., University of Birmingham, **2016**.
- [6] G. D. Parfitt, The dispersion of fine particles in liquid media, in: *Mixing in the Process Industries (Ed2)*, Elsevier (ISBN 0408115742), **1997**.
- [7] U. Teipel, I. Mikonsaari, S. Torry, Wettability Analysis, in: *Energetic Materials*, Wiley, **2005**, pp. 403–431.
- [8] D. Ramirez, J. Kalman, Influence of htpb variants on the wettability of ammonium perchlorate, *AIAA Scitech 2020 Forum* **2020**, 1 PartF, 1–15, doi: 10.2514/6.2020-0425.
- [9] Charles Ross & Son Company, "How to choose the right mixer for high-viscosity mixing applications.," **2020**.
- [10] D. V. Nance, *An Examination of the Resonant Acoustic Mixer's Flow Field (AFRL-RW-EG-TR-2013-108)*, Defense Technical Information Center, **2013**.
- [11] Resodyn, "Resonance for power and efficiency acoustics for performance and quality," **2019**.
- [12] P. Lucon, G. Sperry, J. Whaley, RAM mixing of liquids and pastes, in: *Resodyn Technical Interchange*, Butte, Montana, USA, **2016**.
- [13] R. Manasseh, Acoustic bubbles, acoustic screaming, and cavitation microstreaming, in: *Handbook of Ultrasonics and Sono-Chemistry*, Springer Singapore, **2016**, pp. 33–68.
- [14] H. Howe, J. Warriner, A. Cook, S. Coguill, L. Farrar, *Apparatus and Method for the Resonant Vibratory Mixing US Patent 7188993 B1*, **n.d.**
- [15] H. Howe, J. Warriner, A. Cook, S. Coguill, L. Farrar, *Method for Resonant Vibratory Mixing US Patent 7866878 B2*, **n.d.**
- [16] "Resodyn Acoustic Mixers, Inc., How RAM Mixes - Faraday," **n.d.**, available at: <https://resodynmixers.com/how-ram-mixes/#faraday> (accessed: 9th June 2021).
- [17] "Resodyn Acoustic Mixers, Inc., How RAM Mixes - Viscous," **n.d.**, available at: <https://resodynmixers.com/applications/viscous/> (accessed: 9th June 2021).
- [18] D. V. Nance, *Simulating the Resonant Acoustic Mixer, AFRL-RW-EG-TR-2013-079*, **2013**.
- [19] A. J. Claydon, D. P. P. Gill, D. G. Kister, M. S. Gaultier, D. N. Flood, Resonant Acoustic Mixing of Polymer Bonded Explosives., Cranfield Defence and Security, **2021**.
- [20] E. L. McCloy, Resonant Acoustic Mixing: Pushing the Boundaries. MSc Thesis, Cranfield University, **2016**.
- [21] P. Lucon, Mixing with Vacuum Assist, in: *Resodyn Technical Interchange*, Butte, Montana, USA, **2016**.
- [22] S. L. Coguill, Z. R. Martineau, Vessel Geometry and Fluid Properties Influencing Mix Behaviour of Resonant Acoustic Mixing, in: *38th*

- International Pyrotechnic Seminar*, Denver, CO, USA, **2012**.
- [23] R. Lopez, J. McCarthy, Modelling of a Resonant Acoustic Mixer Using the Lattice Boltzmann Method with a Free Surface Coupled with the Discrete Element Method (419d), in: *AIChE Annual Meeting*, **2018**.
- [24] R. Tanaka, N. Takahashi, Y. Nakamura, Y. Hattori, K. Ashizawa, M. Otsuka, Verification of the mixing processes of the active pharmaceutical ingredient, excipient and lubricant in a pharmaceutical formulation using a resonant acoustic mixing technology, *RSC Adv.* **2016**, *6*, 87049–87057, doi: 10.1039/c6ra16209f.
- [25] Resodyn, “Innovative Mixing Solutions,” **2020**.
- [26] Sutton G, *Rocket Propulsion Elements (Ed 9) (978-1-118-75365-1)*, John Wiley & Sons, New Jersey, **2017**.
- [27] R. Muthiah, B. R. Gupta, V. N. Krishnamurthy, Rheology of HTPB propellant. I. Effect of solid loading, oxidizer particle size, and aluminum content, *J. Appl. Polym. Sci.* **1992**, *44*, 2043–2052.
- [28] D. M. Kalyon, S. Aktaş, Factors Affecting the Rheology and Processability of Highly Filled Suspensions, *Annu. Rev. Chem. Biomol. Eng.* **2014**, *5*, 229–254, doi: 10.1146/annurev-chembioeng-060713-040211.
- [29] M. Kohga, Y. Hagihara, Rheology of concentrated AP/HTPB suspensions prepared at the upper limit of AP content, *Propellants, Explos. Pyrotech.* **2000**, *25*, 199–202, doi: 10.1002/1521-4087(200009).
- [30] A. K. Mahanta, M. Goyal, D. D. Pathak, Rheokinetic analysis of hydroxy terminated polybutadiene based solid propellant slurry, *E-Journal Chem.* **2010**, *7*, 171–179, doi: 10.1155/2010/750393.
- [31] A. Singh, C. Ness, R. Seto, J. J. de Pablo, H. M. Jaeger, Shear Thickening and Jamming of Dense Suspensions: The “Roll” of Friction, *Phys. Rev. Lett.* **2020**, *124*, 248005, doi: 10.1103/PhysRevLett.124.248005.
- [32] R. Lade, K. Wasewar, R. Sangtyani, A. Kumar, D. Shende, D. Peshwe, Effect of aluminum nanoparticles on rheological behavior of HTPB-based composite rocket propellant, *J. Energ. Mater.* **2019**, *37*, 125–140, doi: 10.1080/07370652.2018.1543737.
- [33] R. Lade, K. Wasewar, R. Sangtyani, A. Kumar, D. Shende, D. Peshwe, Rheological and wall-slip behaviour of composite propellant suspension containing Al-nanopowder, *J. Energ. Mater.* **2018**, *36*, 468–484, doi: 10.1080/07370652.2018.1493056.
- [34] J. R. Brodier, The Benefits and Constraints of Mixing Nanometric Material with Resonant Acoustics. MSc Thesis, Cranfield University, **2017**.
- [35] L. T. Deluca, Innovative solid formulations for rocket propulsion, *Eurasian Chem. J.* **2016**, *18*, 181–196, doi: 10.18321/ectj424.
- [36] G. Ondrey, Resodyn unveils “entirely new” mixing technology., *Chem. Eng.* **2007**, *114*, 13.
- [37] M. R. Andrews, C. Collet, A. Wolff, C. Hollands, Resonant Acoustic® Mixing: Processing and Safety, *Propellants, Explos. Pyrotech.* **2020**, *45*, 77–86, doi: 10.1002/prop.201900280.
- [38] North Atlantic Treaty Organisation, *Standardization Agreement (STANAG) 4170 - Principles and Methodology for the Qualification of Explosive Materials for Military Use (Ed2)*, **2008**.
- [39] M. Zebregs, A. E. H. J. Mayer, A. E. D. M. van der Heijden, Comparison of Propellant Processing by Cast-Cure and Resonant Acoustic Mixing, *Propellants, Explos. Pyrotech.* **2020**, *45*, 87–91, doi: 10.1002/prop.201900169.
- [40] A. P. Nelson, T. A. Cross, Processing Benefits of Resonance Acoustic Mixing on High Performance Propellants and Explosives, *Proc. Int. Pyrotech. Semin.* **2012**, *38th*, 190–206.
- [41] A. Vandenberg, K. Wille, Evaluation of resonance acoustic mixing technology using ultra high performance concrete, *Constr. Build. Mater.* **2018**, *164*, 716–730, doi: 10.1016/j.conbuildmat.2017.12.217.
- [42] N. Rumeau, D. Threlfall, A. Wilmet, ResonantAcoustic® Mixing – Processing and Formulation Challenges for Cost Effective Manufacturing, *Insensitive Munitions Energ. Mater. Technol. Symp.* **2015**, 1–10.
- [43] J. H. Park, K. T. Bae, K. J. Kim, D. W. Joh, D. Kim, J. ha Myung, K. T. Lee, Ultra-fast fabrication of tape-cast anode supports for solid oxide fuel cells via resonant acoustic mixing technology, *Ceram. Int.* **2019**, *45*, 12154–12161, doi: 10.1016/j.ceramint.2019.03.119.
- [44] D. H. Leung, D. J. Lamberto, L. Liu, E. Kwong, T. Nelson, T. Rhodes, A. Bak, A new and improved method for the preparation of drug nanosuspension formulations using acoustic mixing technology, *Int. J. Pharm.* **2014**, *473*, 10–19, doi: 10.1016/j.ijpharm.2014.05.003.
- [45] R. R. Nellums, B. C. Terry, B. C. Tappan, S. F. Son, L. J. Groven, Effect of solids loading on resonant mixed Al-Bi<sub>2</sub>O<sub>3</sub> nanothermite powders, *Propellants, Explos. Pyrotech.* **2013**, *38*, 605–610, doi: 10.1002/prop.201300038.
- [46] D. Jubb, Resonant Acoustic Mixing (RAM) of energetic materials, *Off. J. Inst. Explos. Eng.* **2017**, 28–31.
- [47] D. Lagrange, D. Howard, E. Miklaszewski, R. Nellums, J. Kosak, Resonant Acoustic Mixing of MTV, in: *SERDP & ESTCP Symposium. Resonant Acoustic Mixing of MTV*, **2018**.
- [48] A. Nelson, M. Miller, “Resonant Acoustic Mixing of High- Energy Composite Materials,” **2018**.
- [49] J. G. Osorio, E. Hernández, R. J. Romañach, F. J. Muzzio, Characterization of resonant acoustic mixing using near-infrared chemical imaging, *Powder Technol.* **2016**, *297*, 349–356, doi: 10.1016/j.powtec.2016.04.035.



- [50] J. G. Osorio, F. J. Muzzio, Evaluation of resonant acoustic mixing performance, **2015**, *278*, 46–56.
- [51] E. Batmaz, K. P. Sandeep, Integration of ResonantAcoustic® mixing into thermal processing of foods: A comparison study against other in-container sterilization technologies, *J. Food Eng.* **2015**, *165*, 124–132, doi: 10.1016/j.jfoodeng.2015.06.013.
- [52] R. Sharma, G. Setia, Mechanical dry particle coating on cohesive pharmaceutical powders for improving flowability - A review, *Powder Technol.* **2019**, *356*, 458–479, doi: 10.1016/j.powtec.2019.08.009.
- [53] K. S. Hope, H. J. Lloyd, D. Ward, A. A. L. Michalchuk, C. R. Pulham, Resonant acoustic mixing: Its applications to energetic materials, *New Trends Res. Energ. Mater.* **2015**, 134–143, doi: 10.13140/RG.2.1.4234.8646.
- [54] M. Puszynski, R. Cook, D. Perkins, G. Cheng, N. Mehta, Using RAM Technology to Mix Primary Explosive Based Composites for Additive Manufacturing, **2018**, 164–174.
- [55] C. M. Yamamoto, E. J. Miklaszewski, J. T. Dunham, A. P. Shaw, N. Surface, C. Division, P. Arsenal, S. Nitrate, Issues and observations during scale up of pyrotechnic flare compositions in resonant acoustic mixers, **2018**, 579–580.
- [56] D. J. Am Ende, S. R. Anderson, J. S. Salan, Development and scale-up of cocrystals using resonant acoustic mixing, *Org. Process Res. Dev.* **2014**, *18*, 331–341, doi: 10.1021/op4003399.
- [57] T. G. Yew, Process Parameters for Resonant Acoustic Mixers (RAM). MSc Thesis, Cranfield University, **2015**.
- [58] R. B. Chavan, R. Thipparaboina, B. Yadav, N. R. Shastri, Continuous manufacturing of co-crystals: challenges and prospects, *Drug Deliv. Transl. Res.* **2018**, *8*, 1726–1739, doi: 10.1007/s13346-018-0479-7.
- [59] Resodyn, “RAM Product Family. Mixing Application Bulletin,” **2020**.
- [60] K. Nagapudi, E. Y. Umanzor, C. Masui, High-throughput screening and scale-up of cocrystals using resonant acoustic mixing, *Int. J. Pharm.* **2017**, doi: 10.1016/j.ijpharm.2017.02.027.
- [61] A. Burn, D. Jubb, WSTC 0151 - RAM applications to production, in: *Weapons Sector Research Framework (WSRF) Annual Conference*, **2020**.
- [62] M. Comet, C. Martin, F. Schnell, D. Spitzer, Nanothermites: A short Review. Factsheet for Experimenters, Present and Future Challenges, *Propellants, Explos. Pyrotech.* **2019**, *44*, 18–36, doi: 10.1002/prop.201800095.
- [63] North Atlantic Treaty Organisation Standardisation Agency, *Allied Ordnance Publication 7 (Ed2). Manual of Data Requirements and Tests for the Qualification of Explosive Materials for Military Use.*, **2003**.
- [64] M. Andrews, C. Collet, C. Hollands, A. Wolff, *Resonant Acoustic Mixing: Qualification Challenges. MSIAC Limited Report L-267*, **2021**.
- [65] V. Dennis, D. J. Flynn, T. Vine, D. Holley, *UK E Smart Certification of Energetics – Phase 1 Final Report Approval Copies*, **2012**.
- [66] Siemens, “Digital Twin | Siemens Global copyright ©Siemens AG,” **2019**.
- [67] A. Rogers, M. Ierapetritou, Challenges and opportunities in modeling pharmaceutical manufacturing processes, *Comput. Chem. Eng.* **2015**, *81*, 32–39, doi: 10.1016/j.compchemeng.2015.03.018.
- [68] R. Singh, A. D. Román-Ospino, R. J. Romañach, M. Ierapetritou, R. Ramachandran, Real time monitoring of powder blend bulk density for coupled feed-forward/feed-back control of a continuous direct compaction tablet manufacturing process, *Int. J. Pharm.* **2015**, *495*, 612–625, doi: 10.1016/j.ijpharm.2015.09.029.
- [69] Certara, “Modern, integrated drug development,” **2020**.
- [70] A. A. L. Michalchuk, K. S. Hope, S. R. Kennedy, M. V. Blanco, E. V. Boldyreva, C. R. Pulham, Ball-free mechanochemistry:: In situ real-time monitoring of pharmaceutical co-crystal formation by resonant acoustic mixing, *Chem. Commun.* **2018**, *54*, 4033–4036, doi: 10.1039/c8cc02187b.
- [71] B. R. Halls, X-Ray Radiography and Fluorescence for Liquid Distribution and Mixing Measurements in Impinging Jet Sprays. PhD Thesis, Iowa State University, **2014**.

