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Proposed Programme of Work at E.R.D.E. on
High Energy Materials for Propellants and Explosives

by

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Approved:

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1.   SUMMARY RECOMMENDATIONS

A modest programme to endeavour to improve the performance of current solid propellants and explosives is proposed.   In the case of solid propellants the aim is to increase delivered specific impulse to about 275 lbf.sec/lbm., and for high explosives to increase the energy by about 70 per cent over Torpex, for use in Service by 1969.

It is possible to make an immediate start on a very small scale, especially in the H.E. field, where the aim is to achieve a 50 per cent increase over Torpex by 1966.

Larger scale work, in up to the 10 lb. scale, on more hazardous systems must await completion of a new facility at E.R.D.E., currently programmed for mid 1966; work has not yet started on construction of this facility, but details have been finalised with M.P.B.W.

This limited programme envisages the use of chemical compounds already known, and which are already available, or can be made available, in the required quantities.   No back-up programme on novel or unknown compounds is proposed; this field remains left to I.C.I. (Nobel) under an A.R.P.A. contract.   It is not proposed to carry out work on propellants or explosives containing beryllium at E.R.D.E.   Work on this field could be justified if A.W.R.E. are able to spare some effort.

Initial effort will be 3 W.P.G. full time, with some additional part time support from other staff for about 2 years; it is proposed that this effort be increased to 8 W.P.G. in mid 1966.   If an additional S.O. can be recruited before then, it should be possible to increase the total W.P.G. effort to 5 or 6.

No additional capital facilities, other than those already approved, are foreseen before 1966/67.   If the work then is successful, additional facilities, or extensive alterations to existing facilities, could be required; this aspect is not discussed in this paper.

2.   SCOPE

This paper concerns a new programme on solid propellants and high explosives; it does not deal with the logical development or modification of existing compositions, since this work does not depend on the use of unusual high energy materials.   For example, the present type of plastic propellant may be modified to improve its size capability, or the use of elastomeric binders in plastic bonded explosives may enable a substitute for RDX/TNT to be developed for shell filling.   Work of this nature is already covered in the E.R.D.E. Research Programme; it is a proposed extension of this which is dealt with in the present paper.   The writers do not discuss the means whereby the required additional staff and money may be found - presumably, if the E.R.D.E. budget and staffing is kept at roughly its present level, then the proposed programme can be undertaken only at the expense of some other work. This policy matter will obviously require separate discussion.

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general, the explosive power will increase in proportion to the square of the
specific impulse, since both are measures of the amount of work the material
can do.   A specific impulse of 254 lbf.sec/lbm. is equivalent to an explosive
power of 133 (taking TNT as 100), which is roughly that of composition B
(60/40 RDX/TNT).   For reasons associated with the mechanism of initiation it
is theoretically possible to make greater gains in energy for a given increase
in hazard with composite materials than is possible with single molecular
species;   this is analogous to the relative safety of high energy composite
propellants in contrast to hazardous mono-propellants such as nitroglycerine.

A high specific impulse, and a high explosive power, both derive from
the generation of a large volume of gaseous products at high temperature.
For some high explosive applications (e.g. underwater) gaseous volume is
unimportant and the only criterion is detonation energy density.   It follows
that materials not suitable for propellant use, because of low product gas
volume, may be useful as underwater, or blast, explosives.

The lines of attack for both propellant and explosive compositions are
self-evident: first, to modify existing compositions by the addition of more
energetic ingredients;   second, to produce new oxidisers and fuels capable,
in theory, of delivering more energy and, third, to devise some new chemical
system for the production of high temperature gaseous products of low molecular
weight (current solid propellants and explosives depend on oxidation reactions -
these might be replaced, for example, by fluoridation).   This order also
represents the degree of complexity and time scale of the project and it is
only in the first field that any practical systems have so far emerged.

In the U.K., with limited technical and financial resources, it is
clearly impractical to explore all these fields with an adequate effort.   A
time scale must also be defined, since it is known that it normally takes
between 5 and 10 years to introduce a new propellant or explosive system into
the Service.   Furthermore, it is desirable to know whether the system under
consideration is required for world-wide military purposes, or whether it will
be utilised solely under special conditions, or for civilian space ventures.
For the latter, much more sensitive and less stable compositions might be
acceptable since they could be handled, stored, and transported in special
environments and could have a limited life.   On the other hand, most military
use implies possible world-wide utility, with a reasonable shelf-life of, say,
3 - 5 years, plus a certain robustness to withstand rough handling and risks
of involvement in fire and fragment attack.   Overall, the cost and availability
of likely materials must be considered, but this may not be of paramount import-
ance since the quantities involved are not likely to be very large.   It must
also be recognised that many of the systems proposed involve the use of ingred-
ients which are themselves toxic, or produce toxic products.   Such systems
might have to be restricted to use in outer space since it seems unlikely that
the risks involved would be acceptable in this country in peacetime for ground
launched military, or civilian, rockets.

Finally, it must be recognised that any new solid propellants developed
are likely to find application only in rocket propulsion, which is a very
limited market.   The costs of the research will not be re-couped by creation
of a widespread civilian use in this country and the prospects of overseas sales
are negligible.   Special high explosives will also find a very limited field of
application.

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compared with 2.7 g/cc for aluminium) so that for volume limited systems, and first stage rockets, it would be unlikely to produce any gain in overall performance.

Elemental beryllium has been studied extensively in the United States and a delivered impulse of about 270 lbf.sec/lbm. (under standard conditions) has been reported for nitrasol systems containing some 8 per cent beryllium. It has a lower density than aluminium (1.8 g/cc) so that the gain is most useful for upper stage rockets. The combustion efficiency tends to be somewhat lower than that of aluminised compositions, but there are no compatibility or stability problems. The two major drawbacks are cost (about £30 a lb. compared with 2/6d. a lb. for aluminium) and extreme toxicity of beryllium and its compounds.

At the present stage in the development of solid propellants it is sometimes possible to achieve an increase in performance by the addition of organic high explosives. This is subject to the limitation previously noted that, due to the mechanism of initiation, an increase in power must be accompanied by an increase in hazard. Over a period of time it has come to be recognised that, with our present standards of safety, we are unlikely to produce an organic molecule with more power than HMX; this has a theoretical specific impulse of 242 lbf.sec/lbm. so it is obvious that the addition of high explosives will not result in very much increase over current performance, except in particular cases such as for slow burning compositions.

It is quite likely that nitramines will, however, play an important part in future propellants since the nitrogen present will evolve as molecular nitrogen, stable up to 4000°K at rocket operating pressures (carbon dioxide and water are appreciably dissociated above 3000°K).

5. NEW OXIDISERS AND FUELS

In view of the limitations mentioned above, it would appear to be essential to consider alternative oxidisers and fuels if substantial gains in impulse are to be achieved. Much work has been done in this field, but the number of promising compounds is very limited.

Alternative perchlorates are anhydrous hydrazine monoperchlorate, hydrazine monoperchlorate hemihydrate, anhydrous hydrazine diperchlorate, hydrazine diperchlorate dihydrate, and nitronium perchlorate. The sensitivity and reactivity of these compounds varies widely. The hydrated forms are unlikely to be of use in propellants because of compatibility problems caused by the interaction of water and binder; the gain in specific impulse by using hydrazine monoperchlorate in lieu of ammonium perchlorate is low, but all the hydrazine materials are useful in explosives work because they can be used to control and improve the shock sensitivity of composite explosives.

Nitronium perchlorate is chemically very reactive and its use will pose severe compatibility problems. The gain in energy is very high; whereas a mixture of aluminium and ammonium perchlorate yields 2273 cal/g., the corresponding aluminium/nitronium perchlorate 3084 yields cal/g., which makes it very attractive in underwater explosive applications. Because of the low product gas volume the "power" and specific impulse are poor but this may be overcome by the addition of hydrogen rich material to the composition. Various

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taken to contain the products of combustion, or to remove unwanted fluorides from them.

7.   THE USE OF BERYLLIUM

Apart from toxicity, the addition of beryllium appears to present no unusual hazard.   It would therefore be technically possible to proceed immediately to the 10 lb. scale for 'K' round evaluation.   However, because of the toxicity risks, it would be impossible to use any existing process building at E.R.D.E.;   a specially designed building would be needed, equipped with an air filtration system.   A small firing emplacement, with air-scrubbers would also be needed.   The total cost of such facilities, on a minimum austere scale, would be very considerable, and certainly require reference to the S.C.C.F.

Assuming such a facility were approved very soon, the earliest it could now be made available would be by mid 1967 - it would not be possible to adapt the already approved remote controlled facility without incurring unacceptable further delays.   But it is doubtful whether it is desirable to put such a toxic risk in an area where a large population resides within a few miles. Appendix I gives some detail of the extreme precautions at N.O.T.S., which is already in a desert with a negligible density of population.   Appendix II gives some detail of recent Thiokol work.   These two brief accounts indicate a severe toxic hazard.

Supposing means were found to surmount this toxicity problem, we must consider what the next steps will be, since there is little doubt that a usable propellant could soon be demonstrated.   It would then be necessary to advance to the 200 lb. mixing scale, for which R.O.F. Bridgwater would not be suitable. Space could probably be found at R.O.F. Bishopton, but the installation of, say, two 200 lb. mixers and associated filling equipment, all fitted with air filtration, would take at least two years to obtain Treasury approval and be installed.   Furthermore, if the propellant were filled into motors, these would have to be fired statically in a special enclosed emplacement with air-scrubbers or, alternatively, the motors would have to be projected into the upper atmosphere at Aberporth or Woomera by something like a "Raven".   Each "shot" would be a very expensive affair.

It is known that A.W.R.E. have experience in handling beryllium and, as they are understood to be short of work, it is possible that they could carry out work on the mixing of beryllium powder with inert binders (such as polyisobutene or polyesters).   This mixture could then be mixed with oxidisers at E.R.D.E., with considerably lower toxicity risk - possibly in an existing building, suitably modified.   However, the problems associated with firing motors, and accidental ignitions would still remain.

Similar arguments can be applied, even more forceably, against the use of beryllium hydride, since here we also have the problem of obtaining enough beryllium hydride to do anything useful, and success is not so clearly assured as with elemental beryllium.   It must be admitted that the overall gain in delivered standard specific impulse is likely to be no more than 8 - 10 per cent over present compositions, accompanied by an inevitable loss in density.   This seems hardly worthwhile, as a gain of about 5 per cent is achievable by development of current compositions associated with the use of charge designs of high loading density and giving less sliver.

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(c) To substitute lithium aluminium hydride, or lithium hydride for aluminium in PAX and Nitranol.

(d) Examine nitramines as possible ingredients.

(e) Prepare and examine nitronium perchlorate as a possible ingredient in either PAX or Nitranol-type compositions.

8.2 Solid Propellants

The first requirement is to obtain sufficient aluminium hydride, lithium aluminium hydride, lithium hydride, various hydrazinium perchlorates and nitronium perchlorate to assess their stability and sensitiveness alone, and in the presence of various binders and/or oxidisers. Preliminary work can start immediately, but on anything over a few ounce scale will have to await completion of the new processing facility. These compounds do not pose any unusual toxic risk; lithium hydride and lithium aluminium hydride are commercially available; hydrazinium perchlorates are not available commercially but could be made on an adequate scale by S.C.E. Branch of E.R.D.E. Nitronium perchlorate is available commercially in the U.S.A., but it is not known whether we could purchase any; it could be synthesised at E.R.D.E., but a fair amount of research and development would be needed. Aluminium hydride is available, in limited quantity, in the U.S., and some may be released to the U.K.

It would be very difficult to synthesise satisfactory material in the U.K., and it would seem that, if the U.S. are not prepared to release material, then this line of work will have to be terminated unless some very promising results are obtained in current U.S. work which would justify a bigger U.K. investment.

It would appear that the initial U.K. programme on both propellants and explosives will occupy 3 W.P.G. full time for about 2 years. After this, the new processing facility will become available and the effort will need to be increased to 8 W.P.G. A further 2 years work should demonstrate whether further effort is required, or whether the field ought to be abandoned. This proposal assumes a continued flow of information from the U.S., together with visits by members of E.R.D.E. staff to appropriate U.S. contractors facilities. If the flow of information ceases, or visits are refused, it will almost certainly be necessary to enlarge the team somewhat.

More British activity in space, or a new military requirement, could alter the situation, although even then it is clear that about 2 years will elapse before any sizeable effort can be deployed owing to the lack of suitable facilities at E.R.D.E.

Thiokol (Elkton Division) have worked on a propellant containing beryllium using a carboxyl-terminated polybutadiene (HC) binder and ammonium perchlorate oxidiser. The binder was cured with MAPO.

Theoretical calculations indicated the optimum formulation to be an 85 per cent total solids, with 13 per cent beryllium, and 2 per cent plasticiser.

An 84 per cent total solids formulation (TP-H-3063) containing 13 per cent beryllium and trimodal ammonium perchlorate was processed easily on the 50-lb. scale. An investigation of the effect on performance of the particle size of beryllium was carried out; some interaction between beryllium and the carboxyl groups in the binder was noticed. Subsequently this was shown to be caused by a halide impurity. Improved results obtained by grinding beryllium in argon and treating surface with a chromium compound.

To reduce toxic hazard the beryllium was pre-mixed with binder (i.e. as we do at E.R.D.E. for aluminium).

Firings of motors carried out in closed tank, with water scrubbing. Efficiencies range from 88 to 92.5 per cent theoretical. Typical standard Isp was 255 or 285 when expanded to 0.50 p.s.i.a. to simulate high altitude firing. (The theoretical Isp is about 285 at sea level condition.)

Apart from toxicity, no unusual risks arose. Impact sensitiveness and stability similar to those of aluminised compositions.

The density of these compositions is around 1.66 gm/cc compared with about 1.85 gm/cc for aluminised compositions with similar total solids loading.

Reference: <u>D.J. Smith et alia.</u> CPIA Publication 18A (Oct. 1963) Bulletin of July 1963 S.P. Meeting. Volume IV.

3. THE NEED FOR HIGH ENERGY SYSTEMS

During the last few years an intensive programme on high energy solid propellants has been carried out in the United States by the Advanced Research Projects Agency (A.R.P.A.). This work was started in an endeavour to achieve a significant improvement in delivered specific impulse and, in the early days, it appeared to be assumed that a notable increase could be achieved by the use of some highly energetic material, either on its own, or in conjunction with existing oxidisers. The current situation is that each successive gain in specific impulse has been dearly bought and to make any further advance recourse must now be made to unusual, or very dangerous, materials. The results of this work have so far been made freely available to the U.K. and this now renders it possible for us to select future lines of work which are most likely to be profitable.

It is understood that, as a matter of policy, A.R.P.A. are eliminating chemistry from their activities and that the chemical work will be taken over by the Service Departments. Some reduction in effort is inevitable since these Departments will probably not be allowed to increase their budgets by the same amount as the reduction in A.R.P.A's expenditure. The changing political situation may also mean that the flow of information may not be as free as it has been in the past unless we show much more activity in this field. Until quite recently the effort devoted to high energy materials in the U.K. has been largely confined to a contract paid for by A.R.P.A. with Messrs. I.C.I. (Nobel Division); this contract is concerned with fluorine compounds and is currently under review pending a decision as to its continuance.

The lack of interest in this country has been due to three main reasons:

    (i)   Absence of a positive project requirement needing a high energy material; this in turn has meant that financial approval for new expenditure in this field would have been difficult to justify.

    (ii) Lack of any suitable facilities at E.R.D.E., or elsewhere, for working with unusually hazardous materials.

    (iii) Theoretical considerations which showed that the chance of reaching the original target was slight.

This situation has been altered recently with the approval given to E.R.D.E. to erect a new process building in which to handle hazardous materials in up to 10 lb. scale and by the acceptance of more modest targets. The new E.R.D.E. facility, which should be in full operation by late 1966, will also be suitable for work on high explosive compositions since, as will be discussed at length later in this paper, several of the compounds now under consideration may have possible applications as ingredients of high explosive composition. Requirements for space research now seem rather more definite and provide an outlet for systems which might not be entirely suitable for military uses. In regard to the possible gain in specific impulse, it is significant that the goal of A.R.P.A. for improved solid propellants has been progressively lowered and is now aimed at a delivered specific impulse (under standard conditions) of 280 lbf.sec/lbm. To date, the highest recorded figures in "real" motors are around 270 lbf.sec/lbm. for a modified double base system containing some 8 per cent beryllium. All propellants must have explosive properties and, in

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At the present stage in planning a clear choice must be made between the introduction of a new propellant or explosive by some definite date and the decision to postpone the nomination of this date for a period, keeping abreast of developments as they arise with the inevitable delay in the introduction of the new store when it is needed.

When there is a clear need for a specific material for a dated project the choice is obvious and a compromise on technical qualities (usually the "third best") has to be made to meet the dated requirement.

This position exists in underwater explosives work; the main design parameters for the new submarine-killer torpedo must be clearly defined by the end of 1966 and subsequent to this date the extent to which changes can be made to the "first generation" design will decrease rapidly with time.

With only two years in hand for at least the major part of the development work it is obvious that the new facility at E.R.D.E. will be too late for the first generation of these weapons and that in the next two years emphasis must be placed on explosives which are partially developed already (PAX, Nitranol, HMX mixed with aluminium or beryllium) and that preparation must be made for the development of successors to these explosives in the new facility for subsequent generations of weapons.

Although there is no dated requirement for propellants it is wise to assume that, with the imminent re-equipment of conventional forces, new propellants will be needed and that the most fruitful line of development will be in the field of tactical, rather than strategic, weapons. Increasing interest in space activities will also demand propellants of improved performance. It is suggested that a programme for the introduction of a new propellant by 1969 is undertaken and that a target specification is laid down as soon as possible.

This paper will not debate the relative merits of solid and liquid systems; suffice to state that there are advantages for solid systems, particularly for military applications, which are of great importance. But the ultimate in chemical energy is achievable by the use of liquid hydrogen with fluorine or oxygen; we are concerned in filling the gap between these systems and current solid propellants.

4.   MODIFICATION OF EXISTING SYSTEMS

This is an obvious line of attack, and the most notable advance in recent years has been the addition of aluminium (in up to 15 per cent concentration) to all types of double base and composite systems. The maximum delivered Isp for plastic propellants has been raised to 250 lbf.sec/lbm.; for polyurethane type to around 245 lbf.sec/lbm. and for modified double base (containing also ammonium perchlorate) to around 255 lbf.sec/lbm. By the use of casting liquids which are almost pure nitroglycerine, and the addition of ammonium perchlorate, the impulse of double base can be raised to about 260 lbf.sec/lbm. but the hazard in manufacture has been appreciably increased.

Continuing on this line, the obvious investigation is to study any other solid light elements. In fact only two remain, namely, lithium and beryllium. Elemental lithium is too reactive to be used in solid propellants without some preliminary treatment. Furthermore it has a very low density (0.53 g/cc

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organic perchlorates have been mentioned, but seem unlikely to have much advantage over ammonium perchlorate.

For fuels, interest centres on compounds with a high proportion of hydrogen. One of the best of these is, of course, CH_2 which is already in use as polyisobutene. Other hydrides are beryllium and aluminium hydrides, lithium hydride, lithium aluminium hydride and lithium borohydride. The boron hydrides have been eliminated by exhaustive U.S. work. Aluminium and beryllium hydrides may be available (in limited quantity and considerable cost) from the U.S.A. and are more stable than was first thought. They are also compatible with some propellant ingredients. However, their density is low and such performance measurements as have been noted are disappointingly low. The other hydrides all appear too reactive to use in contact with oxidisers and will have to be encapsulated before they can be used.

Lithium hydride and lithium aluminium hydride are both commercially available in the U.K., and may find application, particularly in conjunction with fluorine compounds. These materials are very reactive and have to be handled under "dry-box" conditions. In the presence of oxidisers very sensitive mixtures are produced, and it would appear that the most likely use of these compounds would be in a hybrid system in which the "fuel" and "oxidiser" were separated.

Other fuels under consideration are derivatives of acetylene, or azides, or other compounds containing energetic bonds. Such compounds tend to be unstable and/or very sensitive, or incompatible with oxidisers. It seems unlikely that any suitable compounds have been overlooked, but new techniques of preparation and purification could result in compounds becoming available which were hitherto laboratory curiosities. No work in this field is contemplated in the U.K., and the outcome will therefore depend on knowledge of any successes by the U.S. or other friendly countries.

6. THE USE OF SYSTEMS BASED ON FLUORINE

The final alternative is to develop a new combustion system and here interest must centre on fluorine, since most fluorides are thermally very stable. A suitable oxidiser would thus be a solid fluorine compound, used in conjunction with a light element such as lithium, or with a hydride. The fluorine compound itself must be capable of dissociation, and hence much work has gone into the preparation of compounds derived from difluoramine. Such work seems very hazardous and requires special equipment; at this stage it must essentially be a laboratory scale preparative research and must be left with Messrs. I.C.I. Ltd. for as long as their A.R.P.A. contract continues.

Compatibility and sensitiveness problems are likely to arise, nor is it yet certain that the expected gains in performance can be achieved; some thermochemical data have already been revised several times and compounds containing carbon/fluorine bonds appear to have been eliminated from the race. At present nitrogen/fluorine or oxygen/fluorine systems are preferred but clearly fluorine can be used only if it reacts with some other element to form a stable fluoride. Information on the properties of fluorides at high temperatures is sketchy and, in any event, nozzle materials might present a serious technical problem since the expected combustion temperatures will be in the region of $4000^{\circ}K$. Fluorine compounds are frequently toxic, so that special precautions might have to be

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8. CONCLUSIONS AND RECOMMENDATIONS

8.1   Underwater and Air Blast Explosives

The current basic materials are aluminium and ammonium perchlorate. These are bound either in an elastomeric binder (e.g. Nitranol) or a fusible perchlorate binder (e.g. PAX).

The most difficult problem is to obtain a high sensitivity (to shock) coupled with a low sensitiveness (to mild stimuli).   This is a research problem fundamental to the whole programme.

There is a specific problem raised by the underwater torpedo application in that one possible warhead design requires the highest possible combination of underwater blast energy and detonation (C.J.) pressure.   Explosives of the E.D.C.24 type have the highest C.J. pressure but only a moderate blast potential; if the latter is raised by increasing the energy either the reaction rates drop (as with the addition of aluminium) or the sensitiveness becomes unacceptable.   This line of development calls for a nice balance between the various parameters and some progress has been made with a composite modified double base material (F.452/464/19).

Explosive	Detonation Energy cal/g.	Underwater Shock Wave to Pentolite = 1.00	C.J. Pressure Kilobars
RDX/TNT 60/40	1130	1.02	253
E.D.C. 24	1325	1.11	310
E.D.C. 24/Al 73/27	1955	1.35	225
PAX	2444	1.51	very low
Nitranol	2260	1.45	127
F.452/464/19	1735	1.27	275

The programme on explosives (excluding elastomeric and heat resistant types) will therefore be:

(a)   To substitute the hydrazine perchlorates for ammonium or lithium perchlorate in PAX, and to investigate allied problems such as compatibility, corrosion and storage life.

(b)   To substitute pure (or nearly pure) nitroglycerine for cooler plasticisers in Nitranol and to investigate the relation between C-J pressure and under-water blast effect in Nitranol.

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APPENDIX 1

N.O.T.S. TP 3382 Chapter 2, p.37 (September 1963)

Ref. TIL P123252; LS/17115

A mobile propellant facility was completed and is now in operation. This BOONDOCK facility is located approximately 3 miles N.E. of the Salt Wells composite facility and is 1.75 miles from the nearest inhabited building. Area studies indicate that an exclusion radius of 6,500 ft. is adequate for personnel protection in the event of a fire consuming 100 lb. of toxic pro- pellant, if lapse conditions and a wind in excess of 2 m.p.h. exist. Quan- tities of toxic propellant as large as 45 lb. may be processed if steep lapse conditions exist.

Operation of this remote facility has been quite satisfactory for the processing of 8-, 15-, 25-, and 67-lb. beryllium propellant motors.

On 20th July 1963 the first 150-gallon mix was made in the Salt Wells composite facility. A total of 620 lb. of CB-11 propellant containing 68.2 lb. of beryllium was processed.

Air samples were run after the mix and less than 1/100 of the allowable concentration was found in the mix room. On 24th August 1963 the second 150- gallon beryllium mix was made. This mix was also 620 lb. of CB-11 propellant with 68.2 lb. of beryllium. To date a total of approximately 1,720 lb. of beryllium propellants have been processed at N.O.T.S.

The 150-gallon composite facility is operated under tight wind speed and direction restrictions. Operation is carried out on week ends when personnel in the plant can be limited to those necessary for the toxic mix and cast operations. Under these conditions, should an accidental ignition of a large batch of propellant occur, no one will be closer than 10 miles down wind of the mix. The distance is adequate to protect the population from a batch as large as 2,200 lb. of propellant.