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MASTER

November 24, 1980

Lawrence Livermore National Laboratory

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# CONTENTS

# GLOSSARY

| | |
|---|---|
| Banbury intensive mixer | Mixer produced by Farrel-Birmingham Co., Inc., Ansonia-Derby, Conn. |
| Cab-O-Sil grade MS7 | Specific grade of silicon dioxide produced by the Cabot Corp., Boston, Mass. |
| EEMCO rubber mill | Two-roll mill produced by the Erie Engine and Mfg Co., Erie, Pa. |
| HiSil 233 | Specific grade of silicon dioxide produced by Pittsburgh Plate Glass Industries, Inc., Chemical Division, Pittsburgh, Pa. |
| Instron test machine | Universal testing machine produced by the Instron Corporation, Canton, Mass. |
| Killian extruder | Plastics extruder produced by Killian Tool and Mfg. Co., Verona, N.J. |
| L97KVB | Lot of equilibrium polymerized silicone gum produced by Lawrence Livermore National Laboratory. |
| L1668 | Intermediate reinforced silicone gum produced by LLNL. |
| L3219 | Heat-stripped L1668 produced by LLNL. |
| L3223 | Molding compound of L3219, temporary filler, and curing agent or cellular silicon cushion produced from the molding compound by LLNL. Nominal cushion density of 0.63-0.65 $Mg/m^3$. |
| LLNL | Lawrence Livermore National Laboratory, Livermore, Calif. |
| Luperco 101XL | 2,5-dimethyl 2,5-bis(t-butyl peroxyl) hexane at 50% concentration is a curing agent produced by Lucidol Division, Pennwalt Corp., Buffalo, N.Y. |
| $M_N$ | Number average molecular weight. |
| $M_W$ | Weight average molecular weight. |
| MWD | Ratio of $M_W/M_N$. A measure of molecular-weight distribution. |
| Pasadena Hydraulics press | Molding press made by Pasadena Hydraulics, Inc., El Monte, Calif. |

| | |
|---|---|
| SE54 Lot JE 167 | Specific grade and batch of silicone gum produced by General Electric Silicone Products Department, Waterford, N.Y. |
| UCC | Union Carbide Corporation Silicone Rubber, N.Y., N.Y. |
| $V/V_0$ | Ratio of the swollen polymer volume to the original polymer volume. A measure of degree of crosslinking. |
| W97 | Specific grade of silicone gum produced by UCC. |
| Y1587 | Triethoxy-end-blocked dimethylsiloxane fluid produced by UCC. |

# Effect of Reinforcing Grade Silicon Dioxide on the Properties of a Silicone Elastomer and its Cellular Silicone

## ABSTRACT

The addition of reinforcing silicon dioxide to silicone gum has a significant effect on the properties of the elastomer and the cellular silicone cushion made from the elastomer. As the amount of silicon dioxide increases, the hardness, density, and tensile strength of the elastomer increase. Ultimate elongation is less sensitive to the filler content. In the cushion, the stiffness and compression set increase as the filler content increases, but the resiliency of the cushion decreases.

## INTRODUCTION

Lawrence Livermore National Laboratory has used cellular silicones as support materials and space fillers for almost two decades. Until recently, these materials were supplied solely by Union Carbide Corporation (UCC) to its main customer, the weapons complex. Union Carbide no longer makes these materials. Because there were no proven commercial replacements, we had to develop a silicone technology to both synthesize gums and to compound gums to cellular materials.

In the series of compounding steps, silicone gum is mixed with silicon dioxide and a processing aid to create a reinforced gum, L1668. L1668 is bin aged at room temperature for about a month and then heat stripped to a base gum, L3219. L3219 is filled with a temporary filler and catalyzed with a peroxide catalyst to give a molding compound (e.g., L3223). The molding compound is compression die-molded and cured, water washed to remove the temporary filler, dried, and finally post cured to create the product that we are interested in.

A good cellular silicone cushion should have low compression set, high load retention with time, and compatibility with adjacent components. The silicone gum is given its strength by the addition of a reinforcing grade of silicon dioxide. We evaluated the effects of various amounts of reinforcing-grade silicon dioxide (Cab-O-Sil Grade MS7) on the properties of load-deflection and compression set of the cellular product.

## EXPERIMENTAL

### MATERIALS

The silicone gum used for the experiments was General Electric Co.'s SE54, lot JE167. SE54 is a methyl phenyl polysiloxane polymer containing a small number of vinyl groups; it is similar to the UCC's W97 gum, which we used for years. Lot 167 has the following polystyrene-equivalent molecular weights:

$M_W = 745\,500$

$M_N = 333\,500$

$\mathrm{MWD} = 2.24$ .

This $M_W$ is higher than that typical of W97, which varied from 500 000 to 600 000.

Two grades of silicon dioxide were added to the SE54 gum. Cab-O-Sil MS7 was added to reinforce the gum. Pittsburgh Plate Glass's HiSil 233 was added presumably to insure thermal stability during post cure. MS7 grade has a nominal surface area of 200 ± 25 $m^2/g$. UCC's Y1587, ethoxy end-blocked dimethylsiloxane fluid, is also added as a processing aid.

As our starting point, we compounded the following formulation:

| Material | Parts per hundred of gum by wt | wt% |
|---|---|---|
| SE54 gum | 100 | 60.61 |
| MS7 $SiO_2$ | 45 | 27.27 |
| HiSil 233 $SiO_2$ | 6 | 3.64 |
| Y1587 fluid | 14 | 8.48 |

To this original L1668 we added additional quantities of virgin SE54 gum to give the composition series shown in Table 1. The wt% calculations assume no loss of Y1587 with elapsed time and additional milling in of SE54 gum.

In the original series, we formulated TJ119-45 through TJ119-29. TJ119-25 and TJ119-20 were added later.

## COMPOUNDING

The first step in compounding was the preparation of a large batch of TJ119-45 L1668. We prepared about 2 kg of L1668 in five separate batches. For each batch, one-half of the MS7, one-half of the Y1587, and all the gum were mixed in our 1-lb-size Banbury intensive mixer at a blade speed of 45 rpm. Then the other one-half of MS7, one-half of Y1587, and all the HiSil 233 were added and mixing resumed. Total mixing time was 10 min per batch. After the five batches were compounded in the Banbury, they were blended together on our EEMCO two-roll rubber mill. The entire batch was extruded at 77°C through our Killian extruder with its vented barrel, 3.8-cm-diam screw, and an L/D of 24/1. This completed the mixing, broke up any lumps or screened them out, and removed any trash. The approximately 6-mm-thick extruded ribbons of L1668 were bin aged at ambient conditions for at least 31 days.

**TABLE 1. Compositions of L1668s having varying silicon dioxide contents.**

| ID | MS7, pph SE54 by weight | MS7, wt% | HiSil 233, wt% |
|---|---|---|---|
| TJ119-45 | 45 | 27.27 | 3.64 |
| -41 | 41 | 25.75 | 3.44 |
| -38 | 38 | 24.52 | 3.27 |
| -35 | 35 | 23.24 | 3.10 |
| -32 | 32 | 21.88 | 2.92 |
| -29 | 29 | 20.43 | 2.73 |
| -25 | 25 | 18.36 | 2.45 |
| -20 | 20 | 15.51 | 2.07 |

At this point, the other formulations were made from TJ119-45 by adding virgin SE54 gum to L1668 on the rubber mill. The final material was sheeted from the rubber mill and allowed to bin age for 1 week.

The sheets of L1668 were then heat stripped to convert them to L3219. The purpose of the heat-stripping operation was to complete the interaction of silicone polymer with silicon dioxide, to drive off residual Y1587, and to remove any traces of solvent, unreacted cyclic tetramer, and low molecular weight polymer. Stripping was done in an air-circulating oven at 171° to 177°C for 18 h.

All L3219 materials were converted to L3223 molding compounds of the following constant composition:

| Component | Parts by wt | wt% |
|---|---|---|
| L3219 | 100 | 47.50 |
| Ammonium chloride | 110 | 52.26 |
| Luperco 101XL | 0.5 | 0.24 |

The 101XL was added to the L3219 by milling on the rubber mill for 3 min. The ammonium chloride was added to the catalyzed L3219 by mixing in the Banbury for 5 min at a speed of 40 rpm. The L3223 molding compound was sheeted on the rubber mill in a 3-min operation.

The L3223 molding compounds were converted into L3223 cellular silicones by a series of steps. Sheets of molding compound weighing 145 g each were charged to a 20-$cm^2$ × 0.25-cm flat slab steel mold, pressed in a Pasadena Hydraulics press at 20.7 mPa, and cured at 171°C for 10 min under pressure. The press-cured sheets were washed in about 90°C water in a washing machine for seven

1-h cycles, three 2-h cycles, and one 3-h cycle. The cushions were dried at ambient for 16 h and then oven dried at 149°C for 4 h. The dried cushions were post cured for 24 h at 251°C.

## RESULTS

Data obtained on the L1668s are listed in Table 2. It appears that the 7 days of bin aging of the back-blended materials are sufficient. It is surprising that the amount of material extracted by chloroform does not increase as the amount of silicon dioxide decreases. Apparently the 29 parts of MS7 is still sufficient to interact with all of the gum and has done so in the 7 days of bin aging.

For uncured L3219s we measured the weight loss on stripping and the hardness and chloroform extractables. These values are shown in Table 3. Stripping loss is independent of silicon dioxide content and constant at 4%. In L3219 the amount of chloroform extractables does increase as the silicon dioxide content decreases.

For press-cured L3219s we measured hardness before post curing and weight loss on post curing. For fully cured L3219s we measured hardness, density, chloroform extractables, swell ratio and the percent soluble in chlorobenzene. These data are listed in Table 4. Hardness and density decrease as silicon dioxide content decreases. Swell ratio increases as silicon dioxide content decreases. Density and swell ratio are plotted in Fig. 1. The increase in swell ratio could be due to a deficiency of cross linking agent, interference with cross linking at the higher gum levels, or a decrease in the interaction of silicon dioxide and gum. The density curve changes shape at 29 parts of silicon dioxide; this may not be significant. We also measured the tensile properties on dog-bone specimens of all the fully cured L3219s. These data are shown in Table 5 and Fig. 2. The reinforcing action of MS7 silicon dioxide is well illustrated by Fig. 2. Ultimate strength seems to reach a peak at about 35 parts of silicon dioxide. Stress at 100% and 200% elongation increases linearly with silicon dioxide over the range of 25 to

**TABLE 2. Properties of L1668s as a function of time.**

| | TJ119 | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | -45 | -45[a] | -41 | -38 | -35 | -32 | -29 | -25 | -20 |
| **Hardness, Rex O** | | | | | | | | | |
| 0 days | 22 | 11 | 9 | 8 | 7 | 7 | 6 | 5 | 5 |
| 7 days | — | 11 | 10 | 8 | 8 | 8 | 7 | — | — |
| 1 month | 41 | — | — | — | — | — | — | 6 | 5 |
| **Weight loss, %** | | | | | | | | | |
| 7 days | — | 0.14 | 0.14 | 0.12 | 0.15 | 0.15 | 0.17 | — | — |
| 1 month | 0.65 | — | — | — | — | — | — | 0.09 | 0.12 |
| **Chloroform extractables, wt %** | | | | | | | | | |
| 1 day | 47.4 | — | — | — | — | — | — | — | — |
| 7 days | — | 25.4 | 36.6 | 25.7 | 36.1 | 31.0 | 34.8 | — | — |
| 1 month | 34.9 | — | — | — | — | — | — | — | — |
| **Swell ratio** | | | | | | | | | |
| 1 day | 3.50 | — | — | — | — | — | — | — | — |
| 1 month | 3.24 | — | — | — | — | — | — | — | — |

[a] Freshened by remilling.

TABLE 3. Properties of uncured L3219s.

| ID | Hardness, Rex O | Stripping loss, wt% | $CHCl_3$ extractables, wt% |
|---|---|---|---|
| TJ119-45 | 40 | 3.9 | 40.7 |
| -41 | 30 | 4.4 | 42.4 |
| -38 | 25 | 4.1 | 46.0 |
| -35 | 20 | 4.2 | 49.6 |
| -32 | 17 | 4.0 | 50.4 |
| -29 | 15 | 3.9 | 56.3 |
| -25 | 10 | 4.1 | 57.6 |
| -20 | 8 | 4.3 | 50.3 |

TABLE 4. Properties of cured L3219s.

| | Press-cured | | Post-cured | | | | |
|---|---|---|---|---|---|---|---|
| | | | | | | Chlorobenzene soak, | |
| ID | Hardness, Shore A | Loss on post cure, wt% | Hardness, Shore A | Density, $Mg/m^3$ | $CHCl_3$ extractables, wt% | $V/V_0$ | wt% sol. |
| TJ119-45 | 51 | 3.6 | 60 | 1.216 | 2.4 | 3.34 | 3.0 |
| -41 | 49 | 3.5 | 54 | 1.202 | 2.4 | 3.46 | 2.9 |
| -38 | 46 | 3.1 | 48 | 1.189 | 2.6 | 3.59 | 3.0 |
| -35 | 44 | 3.4 | 46 | 1.179 | 2.6 | 3.69 | 3.0 |
| -32 | 42 | 3.0 | 43 | 1.166 | 2.7 | 3.78 | 3.0 |
| -29 | 39 | 3.2 | 41 | 1.155 | 2.7 | 3.96 | 3.2 |
| -25 | 37 | — | 39 | 1.137 | 2.1 | 4.08 | 3.0 |
| -20 | 36 | — | 35 | 1.115 | 3.0 | 4.30 | 3.0 |

TABLE 5. Tensile properties of cured L3219s.

| | | Ultimate tensile strength, kPa | | Ultimate elongation, | Tensile stress, kPa | |
|---|---|---|---|---|---|---|
| ID | n | Av | S | % | 100% elongation | 200% elongation |
| TJ119-45 | 5 | 7295 | 241 | 370 | 1400 | 3082 |
| -41 | 5 | 7405 | 345 | 404 | 1207 | 2737 |
| -38 | 5 | 7729 | 241 | 448 | 1007 | 2351 |
| -35 | 5 | 6929 | 641 | 439 | 889 | 2103 |
| -32 | 5 | 6922 | 490 | 446 | 814 | 1944 |
| -29 | 5 | 6426 | 345 | 448 | 689 | 1655 |
| -25 | 5 | 5123 | 427 | 444 | 552 | 1262 |
| -20 | 5 | 3254 | 572 | 388 | 483 | 1000 |

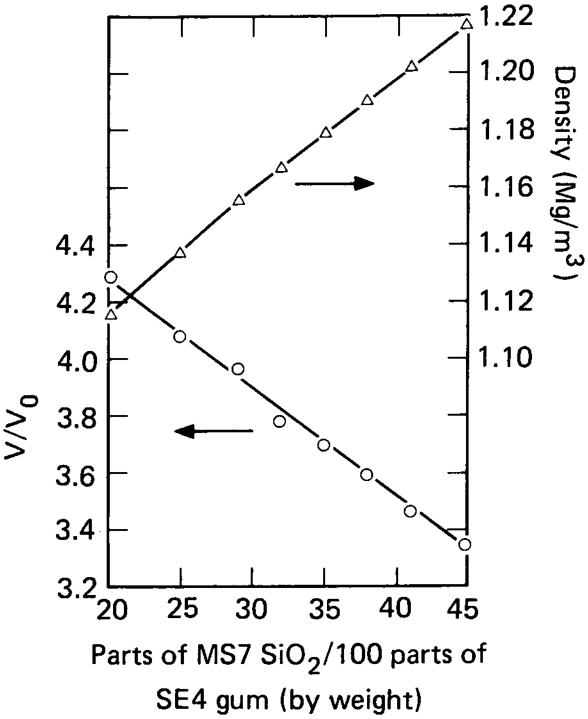

FIG. 1. Effect of silicon dioxide content on density and V/V$_0$ of cured L3219.

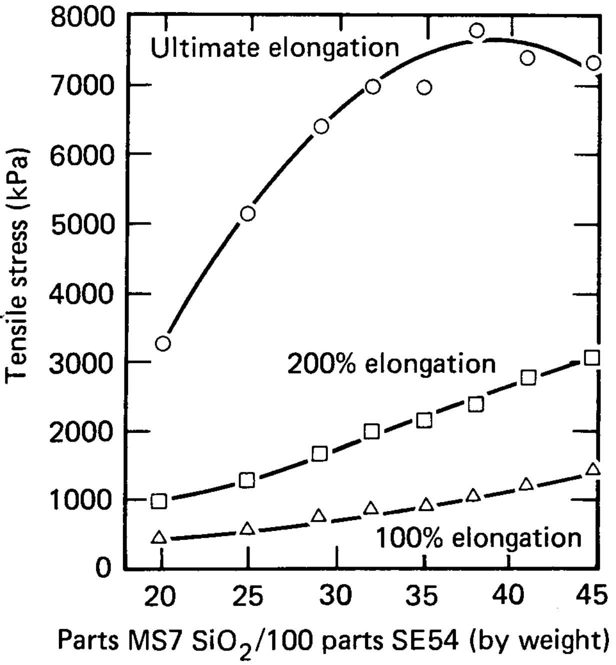

FIG. 2. Effect of silicon dioxide content on the tensile properties of cured L3219.

45 parts. Ultimate elongation is constant over the range of 25 to 38 parts of silicon dioxide.

The L3223 cushions were tested at room temperature to 40% deflection for load-deflection properties on an Instron test machine with a crosshead speed of 1.25 mm/min. The 6.5 cm$^2$ disks were weighed and measured and their densities were calculated. Either 4 or 5 specimens of each L3223 were tested. Third cycle load and unload values at various deflections are listed in Table 6. Unload-to-load ratios, thickness differences between the first and third load cycles, and area differences between the load and unload curves of the third cycle are also calculated. These results and the density measurements are listed in Table 7.

From about 30 parts of silicon dioxide and up, the load increases almost linearly with the silicon dioxide content. Unload values follow the same pattern. In compositions containing 20 to 30 parts of silicon dioxide, the silicon dioxide content has no significant effect on the loading or unloading properties. Apparently the reinforcing effect of silicon dioxide does not begin to dominate the elastomer properties until about 30 parts are present. As the silicon dioxide content increases, the unload-to-load ratio decreases greatly; the hysteresis loss between load and unload curves, and the loss in starting thickness from 1st to 3rd cycle all increase. The addition of silicon dioxide to SE54 gum in increasing amounts produces an increasingly stiffer cushion but also a cushion with decreasing resiliency.

The L3223 cushions were also tested for compression set in our confined-die test. In this test, a 6.35-cm-diam disk of material is compressed 35% in a confined die and placed in an oven at 149°C for 24 h. The sample is cooled to room temperature under compression. Pressure is removed and the height of the sample is measured as a function of time at room temperature. Compression set is calculated from the recovered height at 1 h. Results are shown in Table 8 and % set is plotted in Fig. 3. Compression set decreases as silicon dioxide content decreases and reaches a minimum around 30% silicon dioxide. Ultimate recovery increases as silicon dioxide content decreases and reaches a maximum at 35% silicon dioxide.

TABLE 6. Third-cycle load-deflection properties of L3223s with varying silicon dioxide contents.

| ID | Load, kPa at the following deflections | | | | |
|---|---|---|---|---|---|
| | 5% | 10% | 20% | 30% | 40% |
| TJ119-45 | 44.2 | 70.7 | 117.6 | 203.2 | 565.4 |
| -41 | 38.9 | 63.7 | 105.8 | 179.9 | 514.4 |
| -38 | 35.1 | 58.1 | 95.3 | 162.6 | 439.0 |
| -35 | 29.8 | 50.0 | 82.7 | 138.4 | 382.9 |
| -32 | 26.1 | 44.6 | 74.3 | 124.8 | 344.3 |
| -29 | 23.7 | 41.6 | 70.0 | 116.8 | 305.5 |
| -25 | 23.4 | 42.5 | 72.2 | 119.2 | 298.3 |
| -20 | 21.1 | 39.1 | 68.3 | 118.6 | 323.1 |
| | Unload, kPa | | | | |
| -45 | 20.7 | 38.1 | 67.6 | 122.4 | 554.2 |
| -41 | 20.4 | 37.8 | 65.9 | 116.0 | 475.3 |
| -38 | 21.1 | 38.0 | 64.0 | 109.6 | 387.4 |
| -35 | 19.0 | 34.2 | 57.8 | 97.3 | 331.8 |
| -32 | 17.5 | 32.1 | 54.3 | 91.7 | 314.2 |
| -29 | 18.0 | 32.8 | 54.9 | 91.1 | 275.8 |
| -25 | 18.6 | 35.5 | 60.0 | 97.6 | 273.9 |
| -20 | 17.7 | 33.9 | 57.9 | 97.8 | 278.1 |

TABLE 7. Other properties of L3223s with varying silicon dioxide contents.

| ID | Density, Mg/m$^3$ | Thickness loss, mm | Area difference | Unload-to-load ratio at deflections of | | |
|---|---|---|---|---|---|---|
| | | | | 10% | 20% | 30% |
| TJ119-45 | 0.672 | 0.129 | 291.9 | 0.538 | 0.575 | 0.602 |
| -41 | 0.674 | 0.110 | 237.9 | 0.592 | 0.623 | 0.645 |
| -38 | 0.664 | 0.085 | 166.9 | 0.656 | 0.672 | 0.688 |
| -35 | 0.660 | 0.081 | 149.6 | 0.685 | 0.699 | 0.703 |
| -32 | 0.662 | 0.078 | 107.9 | 0.719 | 0.731 | 0.735 |
| -29 | 0.658 | 0.059 | 91.7 | 0.787 | 0.784 | 0.780 |
| -25 | 0.661 | 0.048 | 82.1 | 0.836 | 0.828 | 0.819 |
| -20 | 0.670 | 0.040 | 89.5 | 0.847 | 0.825 | 0.861 |

TABLE 8. Compression set of L3223s with varying silicon dioxide contents.

| ID | Density, $Mg/m^3$ | Compression, % | % set[a] |  |  | Recovery % of original thickness |  |  |  |  |
|---|---|---|---|---|---|---|---|---|---|---|
|  |  |  | n | Av | S | 2 min | 1 h | 24 h | 7 days | 28 days |
| TJ119-45 | 0.6635 | 35.01 | 5 | 11.98 | 1.39 | 94.19 | 95.80 | 96.77 | 97.01 | 97.30 |
| -41 | 0.6632 | 34.94 | 3 | 10.11 | 0.23 | 95.30 | 96.47 | 97.18 | 97.57 | 97.86 |
| -38 | 0.6558 | 34.99 | 5 | 9.28 | 0.62 | 94.91 | 96.75 | 97.63 | 97.92 | 97.98 |
| -35 | 0.6495 | 35.01 | 3 | 7.97 | 0.32 | 96.21 | 97.34 | 98.19 | 98.44 | 98.70 |
| -32 | 0.6475 | 34.97 | 4 | 6.84 | 0.46 | 96.74 | 97.61 | 98.09 | 98.43 | 98.58 |
| -29 | 0.6471 | 35.03 | 5 | 3.85 | 1.74 | 97.68 | 98.65 | 99.09 | 99.36 | 99.52 |
| -25 | 0.6494 | 35.16 | 3 | 4.67 | 1.27 | 97.78 | 98.36 | 98.75 | 98.71 | 98.79 |
| -20 | 0.6596 | 35.10 | 4 | 6.18 | 0.92 | 96.57 | 97.84 | 97.98 | 98.21 | 98.53 |

[a] Calculated at 1 h recovery from

$$\frac{100 - \%\ \text{recovery at 1 h}}{\%\ \text{compression}} \times 100 = \%\ \text{set} .$$

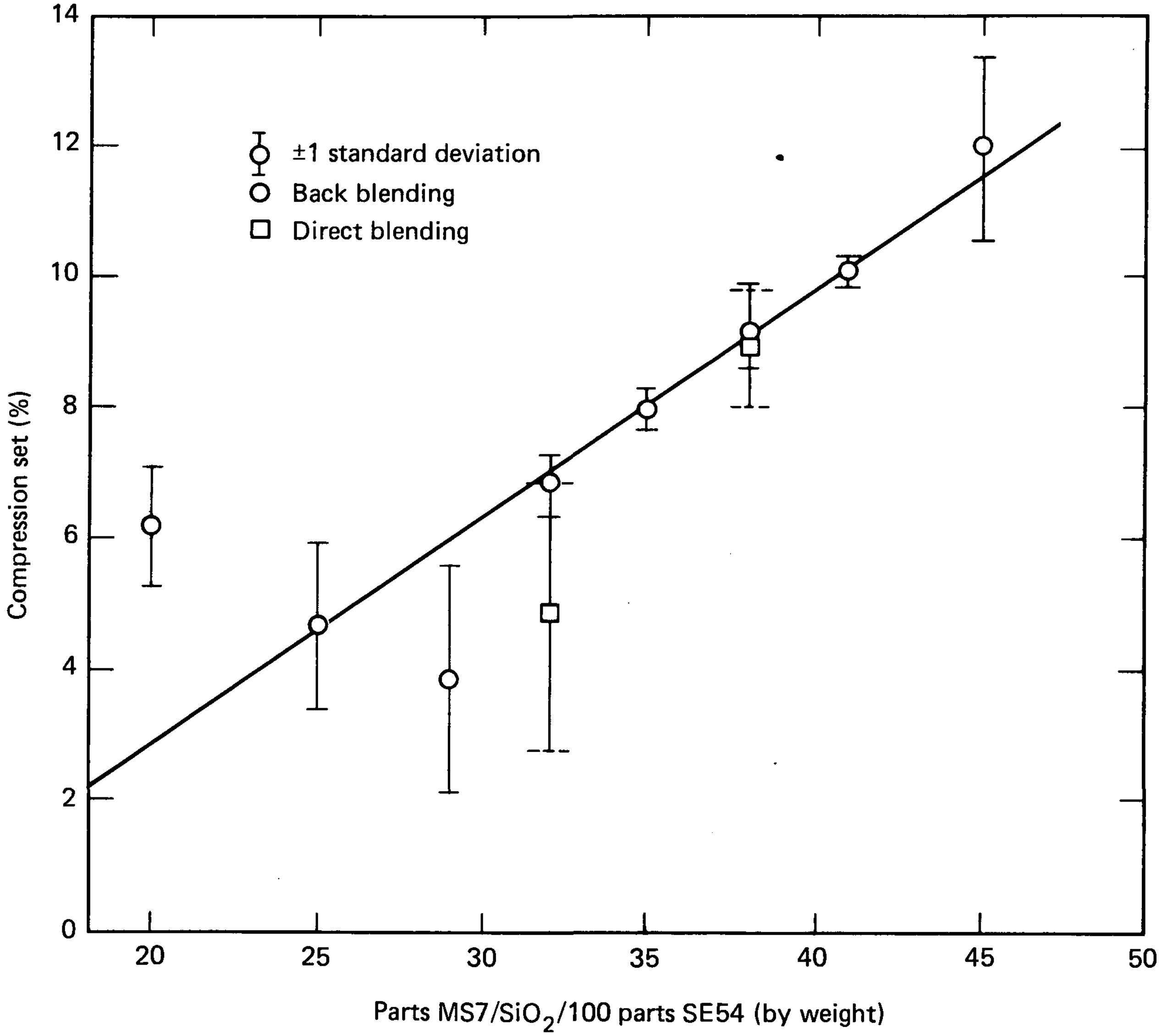

FIG. 3. Confined compression-set as a function of MS7 content.

# COMPARISON WITH DIRECT BLENDING

We selected the back-blending technique to evaluate the effect of MS7 silicon dioxide on the properties of cured elastomers and cellular silicones because it required less compounding effort, and we hoped that it would give us more consistent data than would separate and complete compounding at each silicon dioxide level starting from the gum stage. Direct blending is the normal procedure. We were able to compare direct blending with back-blending at the levels of 32 and 38 parts of MS7. Data for the L3219s are compared in Table 9. The hardness of uncured L3219 is much greater for direct blending than for back-blending. This is probably because of the freshening of the back-blended L1668 when it is remilled to incorporate the additional SE54 gum and the fact that it is then aged only 1 week. The back-blended L3219s have more chloroform extractables than the direct blended. The additional SE54 gum did not react with the silicon dioxide to the extent that the original gum did and so is more soluble in chloroform. At 32 parts of MS7, the blends are alike except that the swell ratio of the back blend is higher than that of the direct blend, probably for the same reason that there are more chloroform extractables. At 38 parts of MS7, the direct blend has a lower swell ratio, ultimate tensile strength, and ultimate elongation.

The load-deflection data for the two blending techniques and the two levels of MS7 are compared in Table 10. At both levels of MS7, back blending produced a softer and more resilient cushion than did direct blending. This again is the diluting effect of virgin SE54 gum on silicon dioxide reinforced SE54 gum.

Confined compression set data are listed in Table 11. Set is also plotted in Fig. 3. At both levels of MS7 the blending techniques have no effect on compression set or ultimate recovery.

**TABLE 9. Properties of L3219s produced by direct blending and by back blending to the same levels of MS7 silicon dioxide.**

| | 32 parts MS7 | | 38 parts MS7 | |
|---|---|---|---|---|
| | Back blending TJ119-32 | Direct blending TJ115 CBA | Back blending TJ119-38 | Direct blending TJ113 |
| Uncured L3219 | | | | |
| Hardness, Rex O | | | | |
| before stripping (L1668) | 8 | 26 | 7 | 41 |
| after stripping (L3219) | 17 | 50 | 25 | 51 |
| $CHCl_3$ extractables, wt% | 50.4 | 41.4 | 46.0 | 27.0 |
| Cured L3219 | | | | TJ140 CBA |
| Hardness, Shore A | 42 | 42 | 46 | 48 |
| Post cured L3219 | | | | |
| Hardness, Shore A | 43 | 43 | 48 | 55 |
| Loss on post cure, wt% | 3.0 | 3.0 | 3.1 | 2.5 |
| $V/V_0$ | 3.78 | 3.23 | 3.59 | 3.40 |
| $CHCl_3$ extractables, wt% | 2.7 | 2.8 | 2.6 | — |
| Density, $Mg/m^3$ | 1.166 | 1.172 | 1.189 | 1.197 |
| Ultimate tensile strength, kPa | | | | |
| n | 5 | 5 | 5 | 5 |
| Av | 6922 | 6736 | 7729 | 6164 |
| S | 490 | 228 | 241 | 159 |
| Ultimate elongation, % | 446 | 416 | 448 | 368 |

TABLE 10. Load-deflection properties of L3223s produced by direct blending and by back blending to the same levels of MS7 silicon dioxide.

| | 32 parts MS7 | | | | | | 38 parts MS7 | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Back blending TJ119-32 | | | Direct blending TJ115 CBA | | | Back blending TJ119-38 | | | Direct blending TJ113 | | |
| | n | Av | S | n | Av | S | n | Av | S | n | Av | S |
| Density, $Mg/m^3$ | | | | | | | | | | | | |
| from load disks | 5 | 0.662 | 0.005 | 4 | 0.660 | 0.011 | 5 | 0.664 | 0.003 | 4 | 0.651 | 0.001 |
| from set disks | 4 | 0.648 | 0.003 | 3 | 0.639 | 0.007 | 5 | 0.656 | 0.002 | 3 | 0.641 | 0.001 |
| 3rd cycle Load, kPa | | | | | | | | | | | | |
| 10% deflection | 5 | 44.6 | 0.61 | 3 | 62.8 | 1.65 | 4 | 58.1 | 3.03 | 4 | 61.6 | 0.36 |
| 20% deflection | 5 | 74.3 | 1.13 | 3 | 105.7 | 4.1 | 4 | 95.3 | 4.92 | 4 | 110.9 | 0.68 |
| 30% deflection | 5 | 124.8 | 1.59 | 3 | 180.1 | 12.9 | 4 | 162.6 | 7.39 | 4 | 185.0 | 0.92 |
| 40% deflection | 5 | 344.3 | 14.8 | 1 | 498.2 | — | 4 | 439.0 | 28.3 | 3 | 505.3 | 12.1 |
| 3rd cycle Unload, kPa | | | | | | | | | | | | |
| 10% deflection | 5 | 32.1 | 0.71 | 3 | 41.7 | 0.89 | 4 | 38.1 | 1.64 | 4 | 39.5 | 0.42 |
| 20% deflection | 5 | 54.3 | 1.09 | 3 | 72.1 | 0.52 | 4 | 64.0 | 2.58 | 4 | 69.6 | 0.52 |
| 30% deflection | 5 | 91.7 | 1.33 | 3 | 127.2 | 5.82 | 4 | 109.6 | 5.92 | 4 | 122.1 | 0.71 |
| 40% deflection | 5 | 314.2 | 20.9 | 1 | 442.5 | — | 4 | 387.4 | 32.0 | 3 | 494.7 | 18.5 |
| Unload/load | | | | | | | | | | | | |
| 10% deflection | | 0.719 | | | 0.664 | | | 0.656 | | | 0.593 | |
| 20% deflection | | 0.731 | | | 0.683 | | | 0.672 | | | 0.628 | |
| 30% deflection | | 0.735 | | | 0.707 | | | 0.688 | | | 0.660 | |
| Thickness loss, mm | | 0.078 | | | 0.101 | | | 0.085 | | | 0.122 | |
| Area difference | | 107.9 | | | 201.5 | | | 166.9 | | | 248.1 | |

**TABLE 11. Compression-set properties of L3223s produced by direct blending and by back blending MS7 silicon dioxide to the same levels.**

| | 32 parts MS7 | | 38 parts MS7 | |
|---|---|---|---|---|
| % set[a] | Back blending TJ119-32 | Direct blending TJ115 CBA | Back blending TJ119-38 | Direct blending TJ140 CBA |
| n | 4 | 3 | 5 | 3 |
| Av | 6.84 | 4.83 | 9.28 | 8.94 |
| S | 0.46 | 2.04 | 0.62 | 0.90 |
| Recovery, % of original thickness | | | | |
| at 2 min | 96.74 | 97.87 | 94.91 | 95.48 |
| at 1 h | 97.61 | 98.29 | 96.75 | 96.87 |
| at 24 h | 98.09 | 98.68 | 97.63 | 96.84 |
| at 7 days | 98.43 | 98.62 | 97.92 | 97.42 |
| at 28 days | 98.58 | 98.87 | 97.98 | 97.84 |

[a]Calculated at 1-h recovery.

# CONCLUSIONS

The amount of MS7 reinforcing silicon dioxide in SE54 silicone gum has a significant effect on the properties of the cellular silicon produced from it when it is present at a level of about 30 parts of MS7 per hundred parts of SE54 by weight. Above about 30 parts of MS7, the load at any deflection increases significantly and almost linearly with MS7 content. The unload also increases but at a smaller and less linear rate. The ratio of unload values to load values decreases greatly as the MS7 content increases. Below 30 parts of MS7, the amount of silicon dioxide has small or no effect on the cushion properties. Apparently the reinforcing effect of silicon dioxide does not begin to dominate the elastomer properties until about 30 parts are present. The compression set of the cushion increases linearly with increasing MS7 content about 30 parts. The addition of reinforcing silicon dioxide to SE54 silicone gum begins to have a significant effect upon cushion properties at about 30 parts of MS7. As the MS7 content increases above 30 parts the cushion becomes increasingly and significantly stiffer, compression set increases, and the resiliency of the cushion decreases.

# RECOMMENDATIONS

Inasmuch as a decrease in silicon dioxide content results in decreased compression set (by our confined test) and increased resiliency of the cellular silicone cushion, a decrease in silicon dioxide content may also decrease the long-term compression set.

We suggest that this possibility be explored with our new LLNL-synthesized gums. Our new equilibrium polymerized silicone gum, L97KVB, has so far been reinforced with 32 parts of MS7. We will reduce the MS7 content to 25 parts to achieve a decrease in compression set and an increase in resiliency. This will significantly decrease ultimate tensile strength but retain ultimate elongation of the rubber and will have only a small effect on load-bearing properties at least up to 30% deflection. We will blend directly at 25 parts.

We will try to test both 25 and 32 part MS7 L97KVBs under compression at ambient temperature for extended time periods to evaluate long-term compression set.

# SUMMARY

Varying the amount of reinforcing silicon dioxide added to silicone gum in the range of 30 to 45 parts of silicon dioxide per hundred parts of gum by weight significantly affects the properties of cured elastomers and of the cellular-silicone cushions made from them.

Increasing the silicon dioxide content increases the hardness, the density, and the ultimate tensile strength of the elastomer but has little effect on its ultimate elongation. It also increases the stiffness and set and decreases the resiliency of the cushion.

Back blending of gum into a higher filled system versus direct blending to the same level of silicon dioxide produces a softer and more resilient cushion but has no effect on compression set.

HLL/cj

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