Silicone Lubricant Flushed from Disposable Syringes: Determination by Atomic Absorption Spectrophotometry

J. RICHARD MILLER, JEROME J. HELPRIN,* and J. S. FINLAYSON

Abstract ☐ Amounts of silicone removable from disposable plastic syringes were determined (as silicon) by the use of atomic absorption spectrophotometry. This method revealed that the silicone fluid employed as a lubricant was present in distilled water expressed from 20 to 60 percent of the syringes. As contrasted with the total silicone extractable by dissolving with methyl isobutyl ketone, water-removable silicone appeared to be removed by a mechanical flushing action.

Keyphrases ☐ Silicone lubricant, plastic syringes—analysis ☐ Plastic syringes—water removable silicone ☐ Methyl isobutyl ketone—silicone extraction ☐ Atomic absorption spectroscopy—analysis

The widespread use of disposable syringes in the clinic and laboratory plus recent reports of extraneous materials extracted from plastic syringes (1) and related equipment (2, 3) prompted the authors to measure the amount of silicone lubricant present in such syringes. Preliminary experiments indicated a wide variation among individual syringes, with certain syringes exhibiting relatively high levels of lubricant. The present work was undertaken to substantiate and explain this variation. It was extended to include an investigation of the amounts of silicone carried from these syringes under conditions simulating their normal usage. Since such usage generally consists of injecting aqueous solutions or suspensions, removal of silicone by flushing with water was studied and quantitated.

EXPERIMENTAL

Materials—Syringes used in this study were new, sterile syringes which were purchased by the carton and represented a number of different manufacturing lots. Methyl isobutyl ketone was certified grade.[1] Silicone lubricant was 200 cps. viscosity grade.[2]

Methods—*Extraction of Total Silicone*—Each of a series of syringes, ranging in size from 0.5 to 20 ml., was partially filled[3] with methyl isobutyl ketone. The plunger was then pulled back to the highest graduation, the syringe rotated, and the fluid expelled. This process was carried out four more times. The washings from a given syringe were then pooled for analysis. The values thus obtained were designated total silicone, since it had been demonstrated by analysis of individual sequential washings that after two to four washings, little more silicone could be extracted.

Extraction of Water-Removable Silicone—Each of a second series of 10-ml. syringes was filled with 1 ml. of distilled water. The plunger was then drawn to the 10-ml. graduation and the syringe was rotated so as to distribute the water over the inner surface of the barrel. The water from 10 syringes was expelled into a screw-capped centrifuge tube. This procedure was carried out four more times, the water from each successive washing being expelled into a separate centrifuge tube.

The water phase was extracted by shaking with 1 ml. of methyl isobutyl ketone. The tubes were centrifuged briefly, and the methyl isobutyl ketone was drawn off with a capillary pipet. This extraction procedure was repeated twice and the three volumes of methyl isobutyl ketone were pooled for analysis.

Recovery experiments were carried out using the procedure for measuring water-removable silicone, since it was felt that this would constitute a more demanding test of the technique than would that for total silicone. When known amounts of silicone lubricant were dispersed in the water phase and extracted as described above, the recovery averaged more than 98%.

Analysis by Atomic Absorption—Silicone lubricant was determined as silicon with an atomic absorption spectrophotometer,[4] using a high-temperature nitrous oxide-acetylene flame. The high temperature was required since silicon forms a refractory oxide at lower temperatures. The following instrument parameters were employed: scale expansion, 1 ×; wavelength 2,516 Å; slit number, 3 (which provided a slit width of 0.3 mm. and a bandpass of 2 Å); source, silicone high-brightness hollow cathode. The sample uptake rate was 5.1 ml./min.

A standard curve (Fig. 1) was prepared for each analytical run. This was done by analysis of appropriate dilutions (in methyl isobutyl ketone) of a stock solution made from a weighed sample of silicone lubricant. Although the use of a new standard curve for each analysis eliminated the effect of variability between experiments, the reproducibility of the standard curve was assessed in the following manner. Four consecutive standard curves were

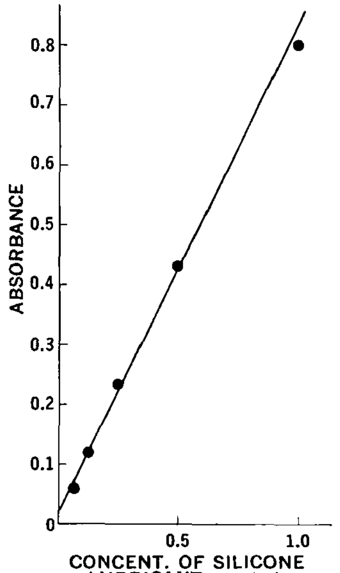

Figure 1—*Typical standard curve for determination of silicone lubricant (as silicon) by atomic absorption spectrophotometry. Dilutions were made in methyl isobutyl ketone; absorbance was measured at 2516 Å.*

[1] Fisher Scientific Co., Fair Lawn, N. J.
[2] 360 Medical fluid, Dow Corning Corp., Midland, Mich.
[3] The volume used per wash was constant for a given size syringe. It was varied according to the capacity of the syringe, and averaged 50% of the graduated volume.
[4] Perkin-Elmer model 303.

Table I—Total Silicone Lubricant Recovered from Plastic Syringes[a]

Syringe Size, ml.	Number of Syringes Tested	Silicone Lubricant Recovered per Syringe, mg. Mean	Range
0.5	90	0.2	<0.1–0.3
1.0	10	2.6	1.2–4.7
2.5	10	0.5	0.3–0.9
5.0	10	3.4	2.8–4.7
10.0	50	7.3	1.3–11.6
20.0	10	6.8	2.5–9.4

[a] Recovered by five extractions with methyl isobutyl ketone; the 0.5-ml. syringes consisted of a glass barrel with a plastic plunger.

Table II—Silicone Lubricant Recovered from Disposable 10-ml. Plastic Syringes by Flushing with Distilled Water[a]

Group	Number of Syringes Tested	Silicone Lubricant (av. mcg./syringe) Water Flush No. 1	2	3	4	5	Sum
1	10	1.5	4.5	1.5	3.0	0.8	11.3
2	10	2.3	2.1	2.1	2.1	2.1	10.7
3[a]	10			None detectable			

[a] These syringes were picked from the same manufacturing lot as Group 2 but exhibited no accumulation of lubricant on the tips of the plungers.

prepared, and the standard deviation of the absorbance value was computed for each concentration of silicone lubricant. The coefficient of variance over the concentration range shown (Fig. 1) averaged 5.6%, corresponding to a relative error of 6.6% in the concentration of silicone over the portion of the curve used for analyses.

Absorbance values of the experimental samples were converted into concentrations of silicone lubricant by using the standard curve. Results were expressed as amount of silicone lubricant per syringe.

RESULTS AND DISCUSSION

Regardless of the size of the syringes, an easily measurable amount of silicone lubricant could be extracted with methyl isobutyl ketone (Table I). That the silicon-containing material in these extracts was indeed silicone lubricant was demonstrated by an IR spectrum of a solution, which was identical with the spectrum of a known silicone lubricant.[2] Analyses of extracts from 10 to 90 individual syringes of each size revealed large variations in the amount of lubricant per syringe (Table I). This variation could not be ascribed to any lack of reproducibility in the analysis, inasmuch as analyses of silicone lubricant solutions used to prepare the standard curve had a standard deviation of only 0.018 mg./ml. at the 0.5 mg./ml. level (*i.e.*, ±3.6% as compared with variations of up to ninefold seen in Table I).

Visual inspection of the rubber tips of syringe plungers under reflected light revealed localized accumulation of a transparent, colorless substance in 20–60% of syringes picked at random. Analyses of these tips alone showed as much as 2.8 mg. of silicone lubricant per tip, or 24% as much as the largest amount found in any syringe (11.6 mg.), although the area of the tip was less than 9% of the graduated area of the barrel of the syringes used. IR analysis of the material from a plunger again showed the characteristic absorption spectrum in the 8- to 12-μ wavelength range which was identical with that of silicone fluid.

In view of these large accumulations of lubricant it appeared possible that silicone might be carried from the syringes by mechanical flushing in the course of injection of aqueous fluids, even though water and silicone lubricant are incompatible (4). For this reason 10-ml. syringes were selected (representing three different manufacturing lots) which displayed the typical accumulations of silicone lubricant on the tips of the plungers, for comparison with syringes of this size in which no lubricant could be seen. These syringes were extracted with distilled water as described under *Methods*. Analyses of the resulting extracts demonstrated the presence of silicone in water expelled from syringes which had visible accumulations of lubricant (Table II). None was detectable in water extracts from syringes without such accumulations (Table II) or from all-glass syringes.

The fact that the amount of lubricant removed by successive water washes was relatively constant (Table II) indicated that it was removed by a mechanical flushing action. A decreasing amount in each successive flush (such as that seen above in successive methyl isobutyl ketone extracts) would have been expected had the lubricant been soluble in water. Furthermore, the sums of the water-removable silicone given in Table II must be regarded as minimum values, since repeated flushing continued to remove appreciable quantities of lubricant.

Proof that the silicon-containing material flushed from syringes with water was silicone lubricant was furnished by the following experiment. A group of 43 plastic syringes (20-ml. capacity) was picked at random from a carton of 50 syringes. Approximately 2 ml. of distilled water was drawn into each syringe, the plunger was withdrawn once to the 20-ml. graduation, the syringe was rotated, and the water was expelled and pooled in a previously unused separator, the stopcock of which was not lubricated. Each syringe was flushed a second time in the same manner, and the washings added to the pool. The water was extracted with two portions (10 ml. each) of methyl isobutyl ketone which were then combined in a porcelain evaporating dish. The funnel was washed with another portion of methyl isobutyl ketone which was added to the previous methyl isobutyl ketone extracts. The dish was heated gently to evaporate the methyl isobutyl ketone extract practically to dryness. The residue in the dish was reconstituted with a few drops of methyl isobutyl ketone, and an IR spectrum was obtained. The identity of this spectrum with that of known silicone lubricant[2] demonstrated that silicone lubricant can be flushed from disposable syringes by handling them in a manner approximating the normal use of such syringes.

Not all syringes of a given size contained detectable amounts of water-removable silicone. The ability of water to remove appreciable quantities of silicone correlated with the presence of local accumulations of silicone lubricant on the plunger tip. However, owing to the translucent nature of plastic syringes, these accumulations are only detectable by visual inspection *after* removal of the plunger from the barrel. Inasmuch as disposable syringes are packaged in a sterile and assembled condition, removal of the plunger for inspection would be impractical for aseptic use.

REFERENCES

(1) M. A. Inchiosa, Jr., *J. Pharm. Sci.*, 54, 1379(1965).
(2) L. Simpson, *Science*, 153, 548(1966).
(3) O. Henry, *ibid.*, 153, 1551(1966).
(4) "Dow Corning 360 Medical Fluid," Dow Corning Corp., Medical Products Div., Midland, Mich., 1966.

ACKNOWLEDGMENTS AND ADDRESSES

Received November 1, 1968, from *Division of Biologics Standards, National Institutes of Health, Bethesda, MD 20014*

Accepted for publication December 23, 1968.

The authors are grateful to Mr. Harry Marshall, Division of Biologics Standards, N.I.H., for performing the IR spectral analyses.

* Deceased.